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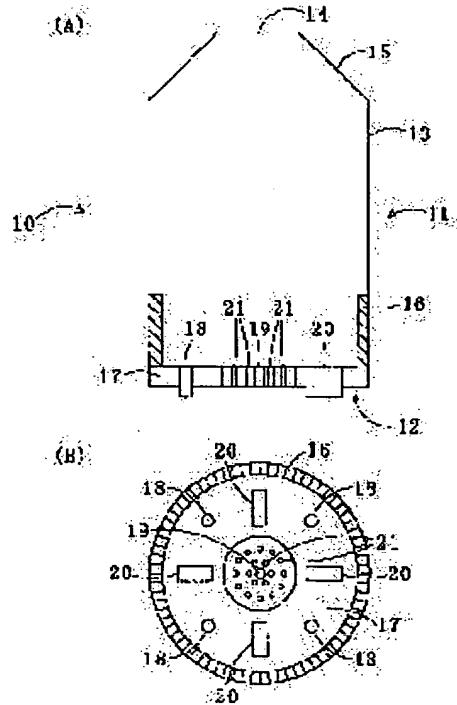
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## (54) FURNACE FOR MANUFACTURING FULLERENE

### (57)Abstract:

PROBLEM TO BE SOLVED: To provide a furnace for manufacturing fullerene in large quantities, economically and simply by a combustion method, by controlling a flowing state of combustion gas.

SOLUTION: The manufacturing furnace 10 provided with a manufacturing furnace main body 11 having feeding ports for a carbon containing compound and oxygen containing gas, manufactures the fullerene in the combustion gas flow produced by burning the carbon containing compound with the oxygen containing gas. Wherein, the mean Reynolds number  $Re$  of the gas flow produced by the combustion is  $0 < Re \leq 2,300$ .



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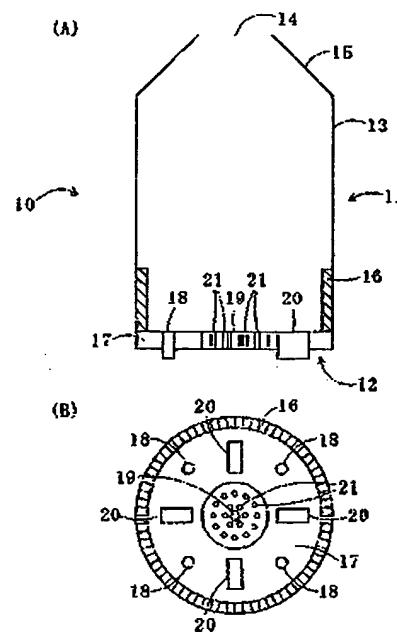
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(54)【発明の名稱】 フラーレンの製造炉

(57)【要約】

【課題】 燃焼法によるフラーレンの製造において、フラーレンの製造炉内の燃焼生成ガスの流れの状態を制御し、フラーレンを大量に且つ安価に、そして簡便に製造することができるフラーレンの製造炉を提供する。

【解決手段】 炭素含有化合物の供給口と酸素含有ガスの供給口が製造炉本体11に設けられ、炭素含有化合物が酸素含有ガスの下で燃焼して燃焼生成ガス流を形成し、燃焼生成ガス流内でフラーレンを製造するフラーレンの製造炉10であって、燃焼生成ガス流の平均レイノルズ数Reが、 $0 < Re \leq 2300$ である。



## 【特許請求の範囲】

【請求項1】 炭素含有化合物の供給口と酸素含有ガスの供給口が製造炉本体に設けられ、該炭素含有化合物が該酸素含有ガスの下で燃焼して燃焼生成ガス流を形成し、該燃焼生成ガス流内でフラーインを製造するフラーインの製造炉であって、前記燃焼生成ガス流の平均レイノルズ数Reが、 $0 < Re \leq 2300$ であることを特徴とするフラーインの製造炉。

【請求項2】 炭素含有化合物の供給口と酸素含有ガスの供給口とを有し製造炉本体に設けられたバーナと、前記炭素含有化合物を前記酸素含有ガスの下で燃焼させて燃焼ガス流が形成される第1反応帯域と、前記第1反応帯域に連続し該第1反応帯域から移入する前記燃焼ガス流中に吹き込み管から主に原料となる炭素含有化合物を供給して燃焼生成ガス流を形成させ、該燃焼生成ガス流内でフラーインが製造される第2反応帯域とを有するフラーインの製造炉であって、前記第2反応帯域内の前記燃焼生成ガス流の平均レイノルズ数Reが、 $0 < Re \leq 2300$ であることを特徴とするフラーインの製造炉。

【請求項3】 請求項1又は2記載のフラーインの製造炉において、前記平均レイノルズ数Reが $0 < Re \leq 1500$ であることを特徴とするフラーインの製造炉。

【請求項4】 請求項1又は2記載のフラーインの製造炉において、前記平均レイノルズ数Reが $0 < Re \leq 1300$ であることを特徴とするフラーインの製造炉。

【請求項5】 請求項1～4のいずれか1項に記載のフラーインの製造炉において、前記炭素含有化合物の供給口と、前記酸素含有ガスの供給口はそれぞれ複数ずつ設けられ、これらの各供給口は各々独立し距離を隔てて前記製造炉本体内に開口していることを特徴とするフラーインの製造炉。

【請求項6】 請求項1～5のいずれか1項に記載のフラーインの製造炉において、前記炭素含有化合物の供給口から供給される炭素含有化合物は予熱されていることを特徴とするフラーインの製造炉。

【請求項7】 請求項1～6のいずれか1項に記載のフラーインの製造炉において、前記酸素含有ガスは不活性ガスを含有することを特徴とするフラーインの製造炉。

【請求項8】 請求項1～7のいずれか1項に記載のフラーインの製造炉において、前記製造炉本体内的圧力が大気圧未満であることを特徴とするフラーインの製造炉。

## 【発明の詳細な説明】

## 【0001】

【発明の属する技術分野】 本発明は、フラーインの製造炉に関する。

## 【0002】

【従来の技術】 フラーインは、ダイヤモンド、黒鉛に次ぐ第三の炭素同素体の総称であり、 $C_{60}$ 、 $C_{70}$ などに代

表されるように6員環と6員環のネットワークで閉じた中空状の炭素分子である。フラーインの存在が最終的に確認されたのは比較的最近の1990年のことであり、比較的新しい炭素材料であるが、その特殊な分子構造ゆえに特異的な物理的性質を示すことが認められ、例えば以下のような広範囲な分野に渡り、革新的な用途開発が急速に展開されつつある。

(1) 超硬材料への応用：フラーインを前駆体として微細結晶粒子をもつ人工ダイヤモンドの製造が可能16なため、付加価値のある耐摩耗材料への利用が期待されている。

(2) 医薬品への応用： $C_{60}$ 、誘導体、光デバイスを用いることで抗癌剤、エイズ・骨粗鬆症・アルツハイマー治療薬、造影剤、ステント材料等の用途としての研究が進められている。

(3) 超伝導材料への応用：フラーイン薄膜に金属性カリウムをドープすると18Kという高い転移温度を持つ超伝導材料をつくり出すことができる事が発見され、多方面から注目を集めている。

20 (4) 半導体製造への応用：レジストに $C_{60}$ を混ぜることでレジスト構造がより一層強化されることを利用し、次世代半導体製造への応用が期待されている。

【0003】 各種炭素素のフラーインの中でも $C_{60}$ 及び $C_{70}$ は比較的の合成が容易であり、それゆえ今後の需要も爆発的に高まることが予想されている。現在知られているフラーインの製造方法としては以下に示す方法が挙げられる。

## (1) レーザ蒸着法

希ガス中に置かれた炭素ターゲットに高エネルギー密度のパルスレーザを照射し、炭素原子を蒸発させてフラーインを合成する方法。電気炉内に不活性ガスが流れる石英管を貫通させ、グラファイト試料をその石英管の中に置く。ガスの流れの上流側からグラファイト試料にレーザを照射し炭素原子を蒸発させると、電気炉出口付近の石英管の内壁に $C_{60}$ や $C_{70}$ などのフラーインを含む膜が付着する。しかし、パルスレーザの1ショット当たりの蒸発量がわずかであり、フラーインの大規模製造には向きである。

## (2) 抵抗加熱法

40 大気圧以下のヘリウムガス雰囲気中でグラファイト棒を通電加熱し、炭素原子を昇華させる方法。グラファイト棒で構成した通電回路での電気抵抗ロスが大きいので、フラーインの大規模製造には向きである。

## (3) アーク放電法

数十kPaに保持したヘリウムガス雰囲気中で2本のグラファイト電極を軽く接触させたり、あるいは1～2mm程度離した状態でアーカ放電を起こし、陽極側の炭素原子を昇華させる方法。現在、工場規模でのフラーインの大規模製造に用いられている。

## 50 (4) 高周波誘導加熱法

高周波誘導により原料グラファイトに過電流を流し、発生するジュール熱により原料グラファイトを加熱して炭素原子を蒸発する方法。

(5) 燃焼法

ヘリウム等の不活性ガスと酸素との混合ガス中でベンゼン等の炭化水素原料を不完全燃焼させる方法。ベンゼン燃料の数%が炭となり、その内の10%程度がフラー  
10レンとなる点で製造効率は良くない。しかし、副燃する煤を液体燃料等に使用可能のこと、製造装置が単純であること等の点で、アーケ合成法に対抗するフラー  
15レンの大置生産法として注目されている。

(6) ナフタレン熱分解法

ナフタレンを約1000°Cで熱分解させる方法。

【0004】以上のように、今までにさまざまなフラー  
10レンの合成法が提案されているが、いずれの方法においても、これまでにフラー  
15レンを安価に大量に製造する方法は確立されていない。これらの方法のうち、最も安価で、効率的な製造方法の一つと考えられるのは燃焼法であり、特表平6-507879号公報には、炭素含有物を火炎中で燃焼させ凝縮物を収集することによるフラー  
20レンの製造方法が記載されている。この製造方法は、炭素含有物を火炎中で燃焼させることによりフラー  
25レンを製造するもので、実質的に燃焼のための燃料とフラー  
30レンの原料は同一の炭素含有物となっている。フラー  
35レンはすす状物質中に含まれて生成されるが、このすす状物質の一部はいわゆるカーボンブラックである。カーボンブラックの製造方法としては、ファーネス法、チャン  
ネル法、サーマル法、アセチレン法などが知られており、工業的な一般的な製造方法としてはファーネス法が挙げられる。この方法では、例えば、円筒状の反応炉を使  
用し、第1反応帯域で炉軸に対して水平方向又は垂直方  
向に空気などの酸素含有ガスと燃料を供給して燃焼ガス流を生じさせ、得られた燃焼ガス流を炉軸方向の下流に設置され第1反応帯域と比較して縮小された断面積を有する第2反応帯域に移動させ、そこで燃焼ガス流中に原  
40料炭化水素(原料油)を供給し反応させてカーボンブラックを生成させる。次いで、カーボンブラックを含有した燃焼ガス流を、第2反応帯域の下流にある第3反応帯  
45域に移動させ、燃焼ガス流に冷却水の噴霧などの冷却処理を行って燃焼ガスを急冷して反応を停止させ、カーボンブラックを回収している。

【0005】

【発明が解決しようとする課題】しかしながら、上記の通常のカーボンブラックの製造方法では、フラー  
10レンはほとんど生成しない。従って、フラー  
15レンの製造においては、得られるすす状物質中に含まれるフラー  
20レンの含有割合をいかに高めるかが、大きな課題となっている。通常、フラー  
25レンの製造は減圧下で行われ、更に、反応  
30領域中に希釈剤を導入する場合もある。これらの減圧度、希釈剤濃度はフラー  
35レンの収率に影響を及ぼすこと

が知られている。そして特表平6-507879号公報には、フラー  
10レンの収率及び組成は火炎中の滞留時間に依存して変化すると記載されており、フラー  
15レンを燃焼法で製造する場合、火炎中の滞留時間を均一に保つことがフラー  
20レンの収率を上げ、組成を一定にすることにつながると考えられる。一般的に、閉じられた容器の中に火炎を形成させると、燃焼反応が活発に行われる火炎中心部の流速は速く、火炎外周部の流速は遅くなる。このため、火炎外周部で上流からの燃焼生成ガスの逆流・巻き込みが起こり、自己循環が発生し易くなる。このような燃焼生成ガスの自己循環は、火炎温度の局所的な高温化を防ぎ、NOxの発生を抑制するという効果がある一方、フラー  
25レンの生成過程においてはフラー  
30レン前駆体の滞留時間の不均一化をもたらす要因ともなる。すなわち、燃焼生成ガスの自己循環が発生すると、燃焼生成ガス中でフラー  
35レンが生成している段階において、この循環する燃焼生成ガス流に乗ったフラー  
40レン前駆体の滞留時間は長くなり、循環する燃焼生成ガス流に巣らなかつたフラー  
45レン前駆体の滞留時間は短くなる。このため、滞留時間の不均一に伴って、フラー  
50レンの収率が低下する。フラー  
55レンは次世代を担う新材料、新素材として多方面から注目されており、このようなフラー  
60レンの滞留時間を制御し、フラー  
65レンを大量に且つ安価に、そして簡便に製造可能となる技術の開発が望まれている。本発明はかかる事情に鑑みてなされたもので、燃焼法によるフラー  
70レンの製造において、フラー  
75レンの製造炉内の燃焼生成ガスの流れの状態を制御し、フラー  
80レンを大量に且つ安価に、そして簡便に製造することが可能なフラー  
85レンの製造炉を提供することを目的とする。

【0006】

【課題を解決するための手段】本発明者らは、燃焼法によるフラー  
10レンの大置かつ安価な製造方法において、フラー  
15レンの製造における原料の最適な燃焼状態を種々検討した結果、フラー  
20レンの製造炉内のフラー  
25レンの生成領域における燃焼生成ガスの流れの状態を制御することで、燃焼生成ガス中のフラー  
30レン前駆体の滞留時間を一様にすることを見出し、本発明を完成させた。前記目的に沿う第1の発明に係るフラー  
35レンの製造炉は、炭素含有化合物の供給口と酸素含有ガスの供給口  
40が製造炉本体に設けられ、該炭素含有化合物が該酸素含有ガスの下で燃焼して燃焼生成ガス流を形成し、該燃焼生成ガス流内でフラー  
45レンを製造するフラー  
50レンの製造炉であって、前記燃焼生成ガス流の平均レイノルズ数Reが、 $0 < Re \leq 2300$ である。

【0007】炭素含有化合物を酸素含有ガスの下で燃焼及び熱分解させて形成した燃焼生成ガス流の平均レイノルズ数Reを、 $0 < Re \leq 2300$ に制御することにより、燃焼生成ガス流内に自己循環流が発生することを防止できる。燃焼生成ガス流内に自己循環流が発生することを防止できると、燃焼生成ガス流内に発生したフラー

レン前躯体の不均一な移動が抑制されて、フーラーレン前躯体の燃焼生成ガス流内での滞留時間を全て一樣とすることができる。

【0008】前記目的に沿う第2の発明に係るフーラーレンの製造炉は、炭素含有化合物の供給口と酸素含有ガスの供給口とを有し製造炉本体に設けられたバーナと、前記炭素含有化合物を前記炭素含有ガスの下で燃焼させて燃焼ガス流が形成される第1反応帯域と、前記第1反応帯域に連続し該第1反応帯域から移入する前記燃焼ガス流中に吹き込み管から主に原料となる炭素含有化合物を供給して燃焼生成ガス流を形成させ、該燃焼生成ガス流内でフーラーレンが製造される第2反応帯域とを有するフーラーレンの製造炉であって、前記第2反応帯域内での前記燃焼生成ガス流の平均レイノルズ数Reが、 $0 < Re \leq 2300$ である。

【0009】第2反応帯域内の燃焼生成ガス流の平均レイノルズ数Reを、 $0 < Re \leq 2300$ に制御することにより、第2反応帯域内での燃焼生成ガス流の自己循環流が発生することを防止できる。燃焼生成ガス流の自己循環流の発生が防止できると、燃焼生成ガス流内に発生したフーラーレン前躯体の不均一な移動が抑制されて、フーラーレン前躯体の第2反応帯域内での滞留時間を一樣とすることができる。

【0010】第1、第2の発明に係るフーラーレンの製造炉において、前記平均レイノルズ数Reが $0 < Re \leq 1500$ であることが好ましい。燃焼生成ガス流の平均レイノルズ数Reを $0 < Re \leq 1500$ とすることにより、燃焼生成ガス流内の自己循環流の発生をより抑えられることがある。このため、燃焼生成ガス流内で形成されたフーラーレン前躯体の燃焼生成ガス流内の滞留時間をより一様とすることができる。

【0011】第1、第2の発明に係るフーラーレンの製造炉において、前記平均レイノルズ数Reが $0 < Re \leq 1300$ であることが更に好ましい。燃焼生成ガス流の平均レイノルズ数Reを $0 < Re \leq 1300$ とすることにより、燃焼生成ガス流内の自己循環流の発生を更に抑えることができる。このため、燃焼生成ガス流内で形成されたフーラーレン前躯体の燃焼生成ガス流内の滞留時間を更に一様とすることができる。

【0012】第1、第2の発明に係るフーラーレンの製造炉において、前記炭素含有化合物の供給口と、前記酸素含有ガスの供給口はそれぞれ複数ずつ設けられ、これらの各供給口は各自独立して能率を高めて前記製造炉本体内に開口していることが好ましい。各自独立して距離を隔てて分散配置された複数の供給口から、それぞれ炭素含有化合物と酸素含有ガスを製造炉本体内に供給することにより、製造炉本体内における燃焼状態をより均一にすることができる、その結果、製造炉本体内の温度を均一化することができる。

【0013】第1、第2の発明に係るフーラーレンの製造

炉において、前記炭素含有化合物の供給口から供給される炭素含有化合物は予熱されていることが好ましい。炭素含有化合物の温度を高めることにより、例えば、炭素含有化合物の自己着火温度以上まで高めることにより、炭素含有化合物の燃焼を均一に行うことができ、製造炉本体内での温度分布をより均一にすることができる。

【0014】第1、第2の発明に係るフーラーレンの製造炉において、前記酸素含有ガスは不活性ガスを含有していることが好ましい。酸素含有ガス中の酸素濃度が不活性ガスにより希釈されて低下するため、製造炉本体内での燃焼を、いわゆる高温空気燃焼状態と類似した状態にすることができる。このため、製造炉本体内の温度はより均一かつ高温にすることができる。ここで、酸素含有ガスの酸素源としては、酸素ガス、空気を使用することができる。また、不活性ガスとしては、ヘリウムガス、アルゴンガス等の不活性ガスを用いる。不活性ガスを用いるため、燃焼時に余分な副製ガスが発生せずフーラーレンの生成には好ましい。フーラーレンの前躯体が形成されてこれがフーラーレンまで成長するのに十分な熱エネルギーが発生すればよいので、酸素含有ガス中における不活性ガスの含有割合には特に制限はない。なお、燃焼火炎温度が高いほどフーラーレンの収率は高くなることが知られている。火炎温度を高める方法としては酸素の添加が有効であるので、フーラーレンの収率という観点からは酸素源としては酸素ガスが好ましい。

【0015】第1、第2の発明に係るフーラーレンの製造炉において、前記製造炉本体内の圧力を大気圧未満とすることが好ましい。製造炉本体内の圧力を大気圧未満にして、炭素含有化合物と酸素含有ガスの混合状態を希薄にすることにより、いわゆる高温空気燃焼と類似した状態を出現させることができるとなる。このため、燃焼が均一に進行して、製造炉本体内の温度はより均一かつ高温にすることができる。

#### 【0016】

【発明の実施の形態】統いて、添付した図面を参照しつつ、本発明を具体化した実施の形態につき説明し、本発明の理解に供する。ここに、図1(A)、(B)はそれぞれ本発明の第1の実施の形態に係るフーラーレンの製造炉の全体概略断面図、炭素含有化合物と酸素含有ガスの供給口の配置を示したバーナの説明図、図2(A)、(B)はそれぞれ本発明の第2の実施の形態に係るフーラーレンの製造炉の全体概略断面図、炭素含有化合物と酸素含有ガスの供給口の配置を示したバーナの説明図である。図1(A)に示すように、本発明の第1の実施の形態に係るフーラーレンの製造炉10は、製造炉本体11と、製造炉本体11の下部に設けられたバーナ12とを有している。以下、これらについて詳細に説明する。製造炉本体11は、円筒形状の側壁部13と、側壁部13の一端側に接続して徐々に外径が縮小して排出口14を形成している端部壁15とを備えている。側壁部13と

端部壁15は、例えばステンレス鋼等の耐熱鋼で構成されている。更に、側壁部13の他端側の内表面には耐火物16がライニングされている。耐火物16としては、例えばアルミナ質の耐火煉瓦やアルミナ質の不定形耐火物を使用することができる。また、排出口14には図示しない排気管の一端側が接続され、他端側は排気ポンプに接続されている。このため、製造炉本体11内を大気圧未満の減圧状態にすると共に、製造炉本体11内で生成したフラーインを含む燃焼生成ガスを製造炉本体11内から外部に排出することができる。

【0017】側壁部13の他端側に取付けられたバーナ12は、例えばステンレス鋼等の耐熱鋼で形成された基盤17と、基盤17に設けられた炭素含有化合物を吐出する複数の供給口18、19と、酸素含有ガスを吐出する複数の供給口20、21とを備えている。図1(B)に示すように、炭素含有化合物の供給口18、19と、酸素含有ガスの供給口20、21は、各自独立に分散して基盤17に設けられている。そして、各供給口18、19、20、21の一方側は製造炉本体11の内側に開口し、他方側は製造炉本体11の外側に開口していずれも図示しない炭素含有化合物の供給管、酸素含有ガスの供給管にそれぞれ接続されている。なお、炭素含有化合物の供給管には加熱器が設けられており、製造炉本体11の内部に吐出される際の炭素含有化合物の温度を、例えば200°Cまで加熱することができる。製造炉本体11の内側に開口している各供給口18、19、20、21の形状は任意であり、略円形、梢円形、三角形や四角形などの多角形状、ひょうたん型などの不定形状であってもよい。本発明者らの知見によれば、円形よりも、長円形や長方形のように長径と短径を持つ形状の方が、酸素含有ガスの加熱や希釈の速度がより速まる。従って、炭素含有化合物の供給口18、19としては、梢円形や略円形が好ましく、酸素含有ガスの供給口20、21としては、スリット状などの長方形状が好ましく、これらを組み合わせるのが特に好ましい。

【0018】炭素含有化合物の供給口18、19と、酸素含有ガスの供給口20、21の配置は、各自独立に製造炉本体11の内側に開口していれば任意とできる。炭素含有化合物の種類や、各供給口18、19、20、21の数などのフラーインの製造炉10の設計条件に合わせていろいろな配置を採用することができるが、例えば、各自の供給口をフラーインの製造炉10の軸心に対して同心円周上に、周方向に交互に配置するならば、製造炉本体11内での燃焼状態がより均一となるので好ましい。この際に、酸素含有ガスの供給口の形状が長径及び短径を持つような場合には、長径から延びた直線が円の中心を通るように配置するのが好ましい。また、製造炉本体11の内側に開口している何れの供給口18、19、20、21も、その開口端部が基盤17の表面と実質的に同一平面上にあっても、突出していてもよいが、

好ましくは実質的に同一平面上がよい。炭素含有化合物の供給口18、19及び酸素含有ガスの供給口20、21から製造炉本体11内に供給される炭素含有化合物及び酸素含有ガスの各流は、各自の供給口18、19、20、21の端部から製造炉本体11内に対して任意の角度で供給してよいが、好ましくは基盤17に対して実質的に垂直となるように、更には、供給される炭素含有化合物及び/又は酸素含有ガスが供給口18、19、20、21の開口端部の中心から実質的に同心円状に拡散するように供給するのが好ましい。

【0019】次に、本発明の第1の実施の形態に係るフラーインの製造炉10を適用したフラーインの製造方法について詳細に説明する。炭素含有化合物の供給口18、19から、炭素含有化合物である炭化水素のガスを、酸素含有ガスの供給口20、21から酸素含有ガスを供給し、これらを燃焼させることで高温の燃焼ガス流を製造炉本体11の下流に向かって発生させる。酸素含有ガスとしては、酸素源である酸素ガスにアルゴンガス等の不活性ガスを任意の割合で混合したガス(例えば、不活性ガスの濃度を0、又は0を超えて90モル%以下の範囲で調整できる)を使用することができる。酸素源としては、フラーインの収率という観点からは酸素ガスが好ましく、酸素源の入手のし易さ等の観点からは空気が好ましい。特に燃焼温度を上げるために、これらの酸素含有ガスは炉内に供給される前に予熱することが好ましい。予熱の方法としては、熱交換器を使用した燃焼生成ガスとの熱交換、いわゆるリジュネレーションバーナ等、公知のいかなる方法を用いても良い。この予熱の温度は常温以上であればいかなる温度でも良いが、フラーインの収率を上げるために極力高温側の方が好ましい。より好ましくは、炭素含有化合物の自己着火温度以上であることが好ましい。炭素含有化合物としては、一酸化炭素、天然ガス、石油ガス等の燃料ガス、重油などの石油系液体燃料、クレオソート油などの石炭系液体燃料を使用することができる。中でもこれらを精製した芳香族系炭化水素を用いることが好ましく、特にベンゼンやトルエン等の芳香族系炭化水素が好ましい。原料の純度は高い方が好ましく、中でも芳香族系炭化水素を用いる際には純度が100%に近いほど好ましい。またフラーインの収率を上げるために、炭素含有化合物も不活性ガス等を用いて希釈することが好ましい。

【0020】統いて、炭素含有化合物が酸素含有ガスの下で燃焼及び熱分解して形成される燃焼生成ガス流について説明する。炭素含有化合物の供給口18、19から供給する炭素含有化合物の量と酸素含有ガスの供給口20、21から供給する酸素ガス量を調整して炭素含有化合物が不完全燃焼する条件で製造炉本体11内に供給すると共に、排出口14に接続された図示しない排出管を介して排気ポンプで製造炉本体11内を大気圧未満、より好ましくは10~300torrの状態に保持して、

図示しない着火手段で炭素含有化合物の燃焼を開始する。ここで、炭素含有化合物と酸素含有ガスは各自独立し距離を隔て分散配置されたそれぞれ複数の供給口18、19、20、21から製造炉本体11内に吐出されるため、製造炉本体11内における燃焼状態を均一にすることができる。また、炭素含有化合物の温度を、例えば、200°Cに高めることにより、炭素含有化合物が均一に燃焼することを助長できる。更に、酸素含有ガス中の酸素ガス濃度はアルゴンガス等の不活性ガスにより希釈されて低下していることに加えて、製造炉本体11内の圧力が大気圧未満となっているため、製造炉本体11内の燃焼状態を高温空気燃焼状態と類似した状態にすることができる。その結果、炭素含有化合物の燃焼が均一に進行して、製造炉本体11内の温度を均一かつ高温(例えば、1000~1900°C、好ましくは1700~1900°C)にすることができる。

【0021】炭素含有化合物の燃焼により製造炉本体11内での燃焼領域の温度は、例えば、1000~1900°C、好ましくは1700~1900°Cの高温になる。このため、未燃焼の炭素含有化合物は容易に熱分解して気化して、炭素含有化合物の燃焼により発生した燃焼ガス内に拡散して燃焼生成ガスを形成する。製造炉本体11内の圧力が大気圧未満で酸素ガス濃度が低い着火状態での燃焼であること、更に、均一燃焼が促進されて燃焼生成ガスの温度が製造炉本体11の軸方向に垂直な方向では実質的に一様になっていることから、燃焼生成ガスにより形成される燃焼生成ガス流内では自己循環流が発生していく。そして、燃焼生成ガスは排出口14から排気ポンプで排気されているので、燃焼生成ガス流はバーナ12から排出口14に向かう一様な流れが主体となる。ここで、燃焼生成ガスの密度は0.01~0.006kg/m<sup>3</sup>、燃焼生成ガスの粘性は5.3×10<sup>-6</sup>~1.4×10<sup>-4</sup>kg/m<sup>2</sup>sec、燃焼生成ガスの平均流速は0.01~4m/sec、製造炉本体11の代表内径の範囲としては0.35~10mと見積もることができる。その結果、燃焼生成ガス流の平均レイノルズ数Reは0<Re≤23000とすることができます。

【0022】平均レイノルズ数Reを0<Re≤2300とすることにより、燃焼生成ガス流内に発生したフラーレン前駆体の不均一な移動が抑制され、フラーレン前駆体の燃焼生成ガス流内の滞留時間を一様とすることができます。その結果、フラーレンの収率を向上させることができることが可能となる。また、製造炉本体11の代表内径と燃焼状態を制御することで、燃焼生成ガスの密度、燃焼生成ガスの粘性、燃焼生成ガスの平均流速をそれぞれ調整して、燃焼生成ガス流の平均レイノルズ数Reを0<Re≤1500、更に、0<Re≤1300とすることができます。平均レイノルズ数Reを0<Re≤1500、更に0<Re≤1300とすることにより、燃焼生

成ガス流内での自己循環流の発生をより抑えて燃焼生成ガス流内で形成されたフラーレン前駆体の燃焼生成ガス流内の滞留時間をより一様とすることができます。その結果、フラーレンの収率をより向上させることができとなる。

【0023】図2(A)に示すように、本発明の第2の実施の形態に係るフラーレンの製造炉22は、製造炉本体23と、製造炉本体23の下部に設けられたバーナ24とを有している。以下、これらについて詳細に説明する。製造炉本体23は、円筒形状の側壁部25と、側壁部25の一端側に接続して徐々に外径が縮小して排出口26を形成している端部壁27とを備えている。側壁部25と端部壁27は、例えばステンレス鋼等の耐熱鋼で構成されている。更に、側壁部25の他端側の内周面には、第1の実施の形態と同様の耐火物16がライニングされている。また、排出口26には図示しない排気管の一端側が接続され、他端側は排気ポンプに接続されている。このため、製造炉本体23内を大気圧未満の減圧状態にすると共に、製造炉本体23内で生成したフラーレンを含む燃焼生成ガスを製造炉本体23内から外部に排出することができる。更に、側壁部25の端部壁27側、及び端部壁27の側壁部25側には、それぞれ主に原料となる炭素含有化合物を製造炉本体23内に吐出する複数の吹き込み管28、29が設けられている。吹き込み管28、29の一端側は製造炉本体23の内側に開口し、他端側には加熱器が設けられた図示しない供給管が接続されており、製造炉本体23の内部に吹き込む主に原料となる炭素含有化合物の温度を、例えば200°Cまで加温することができる。そして、バーナ24と側壁部25の他端側から吹き込み管28が設けられている位置までの側壁部分で囲まれる領域から第1反応帯域が構成されている。また、吹き込み管28が設けられている位置から排出口26までの領域から第2反応帯域が構成されている。

【0024】バーナ24は、例えばステンレス鋼等の耐熱鋼で形成された基盤30と、基盤30に設けられた炭素含有化合物を吐出する複数の供給口31、32と、酸素含有ガスを吐出する複数の供給口33とを備えている。図2(B)に示すように、炭素含有化合物の供給口31、32と、酸素含有ガスの供給口33は、各自独立に分散して基盤30に設けられている。そして、各供給口31、32、33の一側は製造炉本体23の内側に開口し、他方側は製造炉本体23の外側に開口している。更に、図示しない炭素含有化合物の供給管、及び酸素含有ガスの供給管にそれぞれ接続されている。なお、炭素含有化合物の供給管には加熱器が設けられており、炭素含有化合物の供給口31、32から炭素含有化合物が製造炉本体23の内部に吐出される際の温度を、例えば200°Cまで加温することができる。

【0025】製造炉本体23内側に開口している各供給

□31、32、33の形状は任意であり、略円形、梢円状、三角形や四角形などの多角形状、ひょうたん型などの不定形状であってもよい。炭素含有化合物の供給□31、32と、酸素含有ガスの供給□33の配置は、各々独立に製造炉本体23の内側に開口していれば任意である。炭素含有化合物の種類や、炭素含有化合物及び酸素含有ガスの各供給□の数などのフラー-レンの製造炉22の設計条件に合わせていろいろな配置を採用することができるが、例えば、各々の供給□をフラー-レンの製造炉22の中心に対して同心円周上に配置するならば、製造炉本体23内での燃焼状態がより均一となるので好ましい。また、製造炉本体23の内側に開口している何れの供給□31、32、33も、その開口端部が基盤30の表面と実質的に同一平面上にあっても、突出していてもよいが、好ましくは実質的に同一平面上がよい。炭素含有化合物の供給□31、32及び酸素含有ガスの供給□33から製造炉本体23内に供給される炭素含有化合物及び酸素含有ガスの各流は、各々の供給□31、32、33の端部から製造炉本体23内に対して任意の角度で供給してよいが、好ましくは基盤30に対して実質的に垂直となるように、更には、供給される炭素含有化合物及び/又は酸素含有ガスが供給□31、32、33の開口端部の中心から実質的に同心円状に並設するように供給するのが好ましい。

【0026】次に、本発明の第2の実施の形態に係るフラー-レンの製造炉22を適用したフラー-レンの製造方法について詳細に説明する。炭素含有化合物の供給□31、32から、例えば炭化水素ガスを、酸素含有ガスの供給□33から酸素含有ガスを供給し、これらを燃焼させることで高温の燃焼ガス流を製造炉本体23の下流に向かって発生させる。酸素含有ガスとしては、酸素源である酸素ガスにアルゴンガス等の不活性ガスを任意の割合で混合したガス（例えば、不活性ガスの濃度をり、又はりを組んで90モル%以下の範囲で調整できる）を使用することができる。酸素源としては、フラー-レンの収率という観点からは酸素ガスが好ましく、酸素源の入手のし易さ等の観点からは空気が好ましい。特に燃焼温度を上げるため、これらの酸素含有ガスは炉内に供給される前に予熱することが好ましい。予熱の方法としては、熱交換器を使用した燃焼生成ガスとの熱交換、いわゆるリジェネレーションバーナ等、公知のいかなる方法を用いても良い。この予熱の温度は常温以上であればいかなる温度でも良いが、フラー-レンの収率をあげるために極力高温度の方が好ましい。より好ましくは、炭素含有化合物の自己着火温度以上であることが好ましい。

【0027】統いて、供給□31、32から供給された炭素含有化合物が燃焼して形成される燃焼ガス流と、吹き込み管28、29から供給された主に原料となる炭素含有化合物が燃焼ガス流中で熱分解して形成される燃焼生成ガス流について説明する。第1反応帯域では、製造

炉本体23の内部に供給□31、32から供給した炭素含有化合物を、供給□33から供給した酸素含有ガスにより燃焼させて高温の燃焼ガス流を製造炉本体23の下流に向かって発生させる。この第1反応帯域では、高温の燃焼ガスを発生させることが目的であり、その燃焼方法は予混合燃焼、拡散燃焼、層流燃焼、乱流燃焼、高温空気燃焼等、公知のいかなる燃焼方法であってもよい。燃焼は完全燃焼であっても、不完全燃焼であっても良い。しかし、好ましくは第1反応帯域における燃焼は、

10 燃焼に必要な酸素が、理論酸素量以上である、希薄混合気での燃焼の方がよい。酸素含有ガスとしては、酸素源である空気、酸素ガスにアルゴンガス等の不活性ガスを任意の割合で混合したガスを使用することができる。特に高温燃焼におけるNO<sub>x</sub>の発生を抑えるためには、純酸素を使用してもよい。フラー-レンの収率を上げるために、燃焼過程において不活性ガスを用いて希釈することが好ましい。なお、不活性ガスは、酸素含有ガスとしてあらかじめ混合させて供給してもよいが、供給用の専用ノズルから供給しても、供給□31、32から供給される炭素含有化合物に混合させて供給してもよい。

【0028】バー-ナ24から供給する炭素含有化合物としては、一酸化炭素、天然ガス、石油ガス等の燃料ガス、重油、ベンゼン、トルエンなどの石油系液体燃料、クレオソート油などの石炭系液体燃料を使用することができる。特に、炭化水素ガスが好ましい。また、フラー-レン製造時の第1反応帯域における平均温度は、得ようとする目的のフラー-レンによって適宜調整すればよいが、好ましくは1300℃以上、更に好ましくは1600℃以上がよい。これは、燃焼ガスの温度が高温である程、フラー-レンの生産性が上がるからである。なお、温度が高くなり過ぎると、フラー-レンの生産性が落ちる場合がある。また、製造炉本体23内の炉内圧力は大気圧未満であることが好ましく、より好ましい範囲は10～300torrである。

【0029】第2反応帯域では第1反応帯域で形成された燃焼ガス流に吹き込み管28、29から主に原料となる炭素含有化合物を供給し、この炭素含有化合物の一部を燃焼させることによって更に高温にすると共に、残りの炭素含有化合物を熱分解し燃焼ガス流内に拡散させて

40 燃焼生成ガス流を形成して、この燃焼生成ガス流内でフラー-レンを生成させる。主に原料となる炭素含有化合物を部分燃焼させるために、酸素が残存するように第1反応帯域における燃焼を酸素過剰としてもよい。また、第2反応帯域に酸素含有ガスの供給ノズルを設置しても良い。この際、燃焼ガス中に供給される主に原料となる炭素含有化合物は、極力均一に製造炉本体23内に供給されることが好ましい。このため、第2反応帯域に設置する炭素含有化合物の吹き込み管28、29の本数は多いほどよく、また炉内に均等に配置されることが望ましい。また、第2反応帯域の形状も任意であるが、流炉の

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断面形状は変化しないほうが好ましい。その理由は、第1反応帯域と第2反応帯域とで流路の断面形状が変化すると、燃焼ガス流が第2反応帯域に移入する際に流れが乱れ、その結果、主に原料となる炭素含有化合物を供給して形成する燃焼生成ガスの流れも乱れることになって生成するフラーインの収率が低下するためである。フラーインが生成する過程で燃焼生成ガスの流路の断面形状が変化することにより流れの乱れが生じると、生成するフラーインの収率が低下するためである。

【0030】第2反応帯域の平均温度は、製造するフラーインによって適宜選択すればよいが、主に原料となる炭素含有化合物が熱分解し均一に気化して反応するためには、充分高温であることが好ましい。具体的には1000℃以上であることが好ましく、中でも1000～1600℃、特に1700～1900℃であることが好ましい。また、第2反応帯域においては、燃焼生成ガス中の酸素濃度をできるだけ抑制することが好ましい。燃焼生成ガス中に酸素が多量に存在すると、反応帯域すなわち第2反応帯域での主に原料となる炭素含有化合物の一部燃焼が活発に起こり、そのため、反応帯域の不均一が生じることがあるからである。燃焼生成ガス中の酸素濃度は、好ましくは3～10%以下、更に好ましくは0.5～1.5%である。主に原料となる炭素含有化合物としては、従来公知の任意のものを使用することが出来、例えば、ベンゼン、トルエン、キシレン、ナフタレン、アントラセン等の芳香族系炭化水素、クレオソート油、カルボン酸油などの石炭系炭化水素、エチレンヘビーエンドオイル、FCCオイル（流動接触分解残渣油）等の石油系重質油、アセチレン系不飽和炭化水素、エチレン系炭化水素、ペンタンやヘキサン等の脂肪族飽和炭化水素などが挙げられ、これらを単独又は任意の割合で混合して使用してもよい。中でも精製した芳香族系炭化水素を用いることが好ましく、特にベンゼンやトルエン等の芳香族系炭化水素が好ましい。主に原料となる炭素含有化合物の純度は高い方が好ましく、中でも芳香族系炭化水素を用いる際には純度が100%に近いほど良い。

【0031】第2反応帯域の温度は、例えば、1000～1900℃、好ましくは1700～1900℃の高温になっている。このため、第1反応帯域から移入する燃焼ガス流中に供給された主に原料となる炭素含有化合物は容易に熱分解し気化して並散し、燃焼生成ガス流を形成する。製造炉本体23内の圧力が大気圧未満で燃焼ガス濃度が低い、善導状態での燃焼であること、更に、燃焼生成ガスの温度が製造炉本体11の軸方向に垂直方向では実質的に一様になっていることから、燃焼生成ガスにより形成される燃焼生成ガス流内では自己循環流が発生しにくくなっている。そして、燃焼生成ガスは排出口26から排気ポンプで排気されているので、燃焼生成ガス流はバーナ24から排出口26に向かう一様な流れが主

となる。ここで、燃焼生成ガスの密度は0.01～0.006kg/m<sup>3</sup>、燃焼生成ガスの粘性は5.3×10<sup>-5</sup>～1.4×10<sup>-4</sup>kg/m/sec、燃焼生成ガスの平均流速は0.1～4m/sec、製造炉本体23の代表内径の範囲としては0.35～1.0mと見出すことができる。その結果、燃焼生成ガス流の平均レイノルズ数Reは0<Re≤2300とすることができ

【0032】平均レイノルズ数Reを0<Re≤2300とすることにより、燃焼生成ガス流内に発生したフラーイン前駆体の不均一な移動が抑制され、フラーイン前駆体の燃焼生成ガス流内での滞留時間を一様とすることが可能となる。また、製造炉本体23の代表内径と燃焼状態を制御することで、燃焼生成ガスの密度、燃焼生成ガスの粘性、燃焼生成ガスの平均流速をそれぞれ調整して、燃焼生成ガス流の平均レイノルズ数Reを0<Re≤1500、更に、0<Re≤1300とすることができる。平均レイノルズ数Reを0<Re≤1500とすることにより、燃焼生成ガス流内での自己循環流の発生をより抑えて燃焼生成ガス流内に形成されたフラーイン前駆体の燃焼生成ガス流内での滞留時間をより一様とすることが可能となる。その結果、フラーインの収率をより向上させることができ

【0033】以上、本発明の実施の形態を説明したが、本発明は、この実施の形態に限定されるものではなく、例えば、第2反応帯域の長さは、反応炉の大きさ、製造するフラーインの種類などによって適宜選択すればよい。また、図2(A)に示すように、第2反応帯域の位置を第1反応帯域の上部側に設けたが、第2反応帯域は第1反応帯域で形成した燃焼ガス流の流れに対して下流側に逆流して設けられていればよいので、燃焼ガス流の流れ方向を制御することにより、第1反応帯域を取り囲むように第一反応帯域の外側に形成しても、第1反応帯域に取り囲まれるように第1反応帯域の内側に形成してもよい。更に、主に原料となる炭素含有化合物の供給により燃焼生成ガスに乱流などの不規則な流れが発生しないよう制御すれば、吹き込み管の位置は任意に設定することができる。

【0034】【発明の効果】請求項1及びこれに從属する請求項3～8記載のフラーインの製造炉においては、燃焼生成ガス流の平均レイノルズ数Reが、0<Re≤2300であるので、燃焼生成ガス流内でのフラーイン前駆体の滞留時間の均一化が達成でき、フラーインの収率を向上させることができ

【0035】請求項2及びこれに從属する請求項3～8記載のフラーインの製造炉においては、第2反応帯域内の燃焼生成ガス流の平均レイノルズ数Reが、0<Re

$e \leq 2300$ であるので、第2反応帯域内のフラー  
ン前駆体の滞留時間の均一化が達成でき、フラー  
ンの収率を向上させることが可能となる。

【0036】特に、請求項3記載のフラー  
ンの製造炉においては、平均レイノルズ数 $Re$ が $0 < Re \leq 1500$ であるので、燃焼生成ガス流内で形成されたフラー  
ン前駆体の滞留時間がより一様となって、フラー  
ンの収率を向上させることが可能となる。

【0037】請求項4記載のフラー  
ンの製造炉においては、平均レイノルズ数 $Re$ が $0 < Re \leq 1300$ であるので、燃焼生成ガス流内でのフラー  
ン前駆体の滞留時間を更に一様にすることでき、フラー  
ンの収率を更に向上させることが可能となる。

【0038】請求項5記載のフラー  
ンの製造炉においては、炭素含有化合物の供給口と、酸素含有ガスの供給  
口はそれぞれ複数ずつ設けられ、これらの各供給口は各  
々独立し距離を隔てて製造炉本体内に開口しているの  
で、製造炉本体内における燃焼状態を均一化させること  
により製造炉本体内での温度分布を均一にして燃焼生成  
ガス流内に自己循環流が発生することが防止でき、フラー  
ンの生成を効率的に行うことが可能となる。

【0039】請求項6記載のフラー  
ンの製造炉においては、炭素含有化合物の供給口から供給される炭素含有  
化合物は予熱されているので、炭素含有化合物が均一に  
燃焼して製造炉本体内での温度分布をより均一にして燃  
焼生成ガス流内に自己循環流が発生することが防止で  
き、フラー  
ンの生成をより効率的に行うことが可能と  
なる。

【0040】請求項7記載のフラー  
ンの製造炉においては、酸素含有ガスは不活性ガスを含有するので、酸素  
濃度を低下させて燃焼を行うことで製造炉本体内の温度  
をより均一化させることにより燃焼生成ガス流内に自己  
循環流が発生することが防止でき、フラー  
ンの生成を  
効率的に行うことが可能となる。また、酸素濃度を低下  
させて燃焼を行うことで製造炉本体内の温度をより高温

にしてフラー  
ン前駆体の形成を促進でき、フラー  
ンの収率をより高めることが可能となる。

【0041】請求項8記載のフラー  
ンの製造炉においては、製造炉本体内の圧力が大気圧未満であるので、炭  
素含有化合物と酸素が希薄な状態で燃焼反応を進行させ  
ることで製造炉本体内の温度をより均一化させて燃焼生  
成ガス流内に自己循環流が発生することが防止でき、フ  
ラー  
ンの生成を効率的に行うことが可能となる。また、炭素含有化合物と酸素が希薄な状態で燃焼反応を進  
行させることで製造炉本体内の温度をより高温にしてフ  
ラー  
ン前駆体の形成を促進でき、フラー  
ンの収率をより高め  
ることが可能となる。更に、容易に高温が得られることから、純酸素のような高価なガスを用いざと  
も、例えば空気のように入手容易な酸素源を用いてフ  
ラー  
ンの生成が可能となって、フラー  
ン製造における  
コストを大幅に低減することが可能となる。

【図面の簡単な説明】

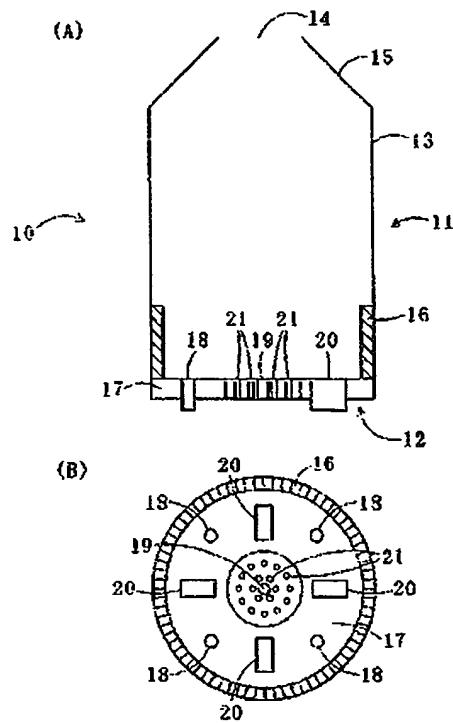
【図1】(A)、(B)はそれぞれ本発明の第1の実施  
の形態に係るフラー  
ンの製造炉の全体概略断面図、炭  
素含有化合物と酸素含有ガスの供給口の配置を示したバ  
ーナの説明図である。

【図2】(A)、(B)はそれぞれ本発明の第2の実施  
の形態に係るフラー  
ンの製造炉の全体概略断面図、炭  
素含有化合物と酸素含有ガスの供給口の配置を示したバ  
ーナの説明図である。

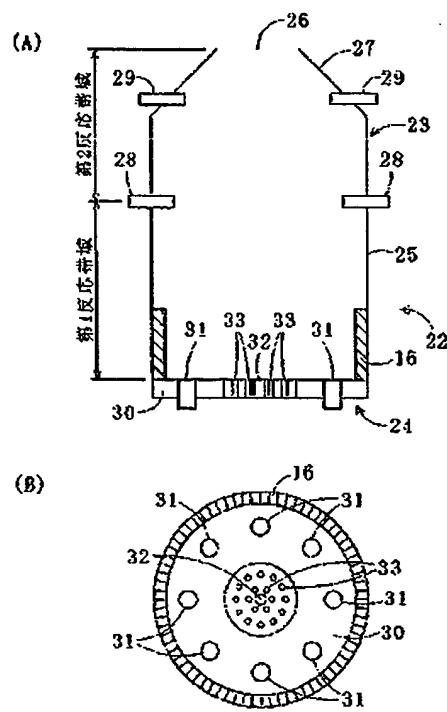
【符号の説明】

10: フラー  
ンの製造炉、11: 製造炉本体、12: バー  
ナ、13: 側壁部、14: 排出口、15: 端部壁、  
16: 耐火物、17: 基盤、18、19: 炭素含有化合物  
の供給口、20、21: 酸素含有ガスの供給口、2  
2: フラー  
ンの製造炉、23: 製造炉本体、24: バ  
ーナ、25: 側壁部、26: 排出口、27: 端部壁、2  
8、29: 吹き込み管、30: 基盤、31、32: 炭素  
含有化合物の供給口、33: 酸素含有ガスの供給口

【図1】



【図2】



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CLAIMS

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[Claim(s)]

[Claim 1] The manufacture furnace of the fullerene characterized by being the manufacture furnace of fullerene which the feed hopper of a carbon content compound and the feed hopper of oxygen content gas are prepared in a manufacture furnace body, and this carbon content compound burns under this oxygen content gas, forms a combustion generation gas stream, and manufactures fullerene within this combustion generation gas stream, and average Reynolds number Re of said combustion generation gas stream being  $0 < Re <= 2300$ .

[Claim 2] The burner which has the feed hopper of a carbon content compound, and the feed hopper of oxygen content gas, and was formed in the manufacture furnace body, The 1st reaction band in which said carbon content compound is burned under said oxygen content gas, and a combustion gas style is formed, Supply the carbon content compound which blows in into said combustion gas style imported from this 1st reaction band succeeding said 1st reaction band, and mainly serves as a raw material from tubing, and a combustion generation gas stream is made to form. The manufacture furnace of the fullerene characterized by being the manufacture furnace of the fullerene which has the 2nd reaction band by which fullerene is manufactured within this combustion generation gas stream, and average Reynolds number Re of said combustion generation gas stream in said 2nd reaction band being  $0 < Re <= 2300$ .

[Claim 3] The manufacture furnace of the fullerene characterized by said average Reynolds number Re being  $0 < Re <= 1500$  in the manufacture furnace of fullerene according to claim 1 or 2.

[Claim 4] The manufacture furnace of the fullerene characterized by said average Reynolds number Re being  $0 < Re <= 1300$  in the manufacture furnace of fullerene according to claim 1 or 2.

[Claim 5] Setting at the manufacture furnace of fullerene given in any 1 term of claims 1-4, the feed hopper of said carbon content compound and the feed hopper of said oxygen content gas are [ two or more ] the manufacture furnace of the fullerene which is prepared and is characterized by for each of these feed hoppers becoming independent respectively, separating distance, and carrying out opening into said manufacture furnace body every, respectively.

[Claim 6] The carbon content compound supplied from the feed hopper of said carbon content compound in the manufacture furnace of fullerene given in any 1 term of claims 1-5 is the manufacture furnace of the fullerene characterized by carrying out the preheating.

[Claim 7] It is the manufacture furnace of the fullerene characterized by said oxygen content gas containing inert gas in the manufacture furnace of fullerene given in any 1 term of claims 1-6.

[Claim 8] The manufacture furnace of the fullerene characterized by the pressure within said manufacture furnace body being under atmospheric pressure in the manufacture furnace of fullerene given in any 1 term of claims 1-7.

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[Translation done.]

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## DETAILED DESCRIPTION

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### [Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the manufacture furnace of fullerene.

[0002]

[Description of the Prior Art] Fullerene is the generic name of the third carbon allotrope which ranks second to a diamond and a graphite, and is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings as represented in C<sub>60</sub>, C<sub>70</sub>, etc. Although it is comparatively that existence of fullerene was finally checked and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.

- (1) Application to a superhard ingredient : since manufacture of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, use to an abrasion resistance material with added value is expected.
- (2) Application to drugs : research as an application of an anticancer agent, an acquired immunodeficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C<sub>60</sub> derivative and an optical device.
- (3) Application to a superconducting material : if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.
- (4) Application to semi-conductor manufacture : it uses that resist structure is further strengthened with mixing C<sub>60</sub> with a resist, and the application to next-generation semi-conductor manufacture is expected.

[0003] Also in the fullerene of various carbon numbers, C<sub>60</sub> and C<sub>70</sub> are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below as the manufacture approach of fullerene learned now is mentioned.

- (1) How to irradiate the pulse laser of a high energy consistency at the carbon target placed into laser vacuum deposition rare gas, evaporate a carbon atom, and compound fullerene. Into an electric furnace, the quartz tube with which inert gas flows is made to penetrate, and a graphite sample is placed into the quartz tube. If laser is irradiated from the upstream of the flow of gas at a graphite sample and a carbon atom is evaporated, the soot containing fullerene, such as C<sub>60</sub> and C<sub>70</sub>, will adhere to the wall of the quartz tube near an electric furnace outlet. However, the evaporation per shot of a pulse laser is slight, and it is unsuitable for extensive manufacture of fullerene.
- (2) The approach to which carry out energization heating of the graphite rod in the gaseous helium ambient atmosphere below resistance heating method atmospheric pressure, and a carbon atom is made to sublimate. Since the electric resistance loss in the energization circuit constituted from a graphite rod is large, it is unsuitable for extensive manufacture of fullerene.
- (3) the-ten number of arc discharge methods -- the approach to which the carbon atom by the side of a

lifting and an anode plate is made to sublimate arc discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium ambient atmosphere held to kPa, or having detached about 1-2mm. It is used for extensive manufacture of the fullerene in current and a works scale.

(4) How to heat an eddy current to raw material graphite by radio frequency heating method high frequency induction, heat raw material graphite with a sink and the Joule's heat to generate, and evaporate a carbon atom.

(5) The approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as combustion method helium, and oxygen. Manufacture effectiveness is not good at the point that several% of a benzene fuel serves as soot, and about 10% of them becomes fullerene. However, the soot which sub\*\* is observed as a method of mass-producing the fullerene which opposes an arc synthesis method in respect of a manufacturing installation being usable to liquid fuel etc., and simple etc.

(6) The approach of carrying out the pyrolysis of the naphthalene thermal decomposition method naphthalene at about 1000 degrees C.

[0004] As mentioned above, although the synthesis method of various fullerene by current is proposed, in which approach, the method of manufacturing fullerene in large quantities cheaply until now is not established. A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, and the manufacture approach of the fullerene by burning a carbon inclusion in a flame in the Patent Publication Heisei No. 507879 [ six to ] official report, and collecting condensates in it is indicated. When this manufacture approach burns a carbon inclusion in a flame, fullerene is manufactured and the fuel for combustion and the raw material of fullerene serve as the same carbon inclusion substantially. Although fullerene is contained in the soot-like matter and it is generated, a part of this soot-like matter is the so-called carbon black. As the manufacture approach of carbon black, the furnace method, a channel process, thermal \*\*, the acetylene method, etc. are learned, and the furnace method is mentioned as the industrial general manufacture approach. By this approach, use a cylinder-like fission reactor, supply oxygen content gas and fuels, such as air, to horizontal or a perpendicular direction to \*\*\*\* in the 1st reaction band, for example, and a combustion gas style is produced. Make it move to the 2nd reaction band which has the cross section which was installed in the lower stream of a river of furnace shaft orientations in the obtained combustion gas style, and was reduced as compared with the 1st reaction band, and supply coal-for-coke-making-ized hydrogen (stock oil), it is made then, to react into a combustion gas style, and carbon black is made to generate. Subsequently, the combustion gas style containing carbon black is moved to the 3rd reaction band on the lower stream of a river of the 2nd reaction band, cooling processing of spraying of cooling water etc. is performed in the style of combustion gas, combustion gas is quenched, a reaction is stopped, and carbon black is collected.

[0005]

[Problem(s) to be Solved by the Invention] However, by the manufacture approach of the above-mentioned usual carbon black, fullerene is hardly generated. Therefore, in manufacture of fullerene, it has been a big technical problem how the content rate of the fullerene contained in the soot-like matter obtained is raised. Usually, manufacture of fullerene is performed under reduced pressure and a diluent may be further introduced all over a reaction field. It is known whenever [ these reduced pressure ] that diluent concentration will affect the yield of fullerene. And when it is indicated that the yield of fullerene and a presentation change to the Patent Publication Heisei No. 507879 [ six to ] official report depending on the residence time in a flame and it manufactures fullerene with a combustion method, maintaining the residence time in a flame at homogeneity gathers the yield of fullerene, and it is considered to lead to carrying out a presentation to regularity. If a flame is made to form into the closed container generally, the rate of flow of the flame core where a combustion reaction is performed actively will be quick, and the rate of flow of the flame periphery section will become slow. For this reason, a back flow and contamination of the combustion generation gas from the upstream happen in the flame periphery section, and it becomes easy to generate self-circulation. Such self-circulation of combustion generation gas prevents local elevated-temperature-ization of flame temperature, and while it is effective in controlling generating of NOx, it also becomes the factor which brings about ununiformity-ization of

the residence time of a fullerene precursor in the generation process of fullerene. That is, if self-circulation of combustion generation gas occurs, the residence time of the fullerene precursor which the residence time of the fullerene precursor which rode on this combustion generation gas stream through which it circulates in the phase which fullerene is generating in combustion generation gas became long, and did not ride on the combustion generation gas stream through which it circulates will become short. For this reason, the yield of fullerene falls in connection with the ununiformity of the residence time. Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and the residence time of such fullerene is controlled, and development of the technique whose manufacture is attained cheaply and simple in large quantities in fullerene is desired. This invention was made in view of this situation, and controls the condition of the flow of the combustion generation gas in the manufacture furnace of fullerene in manufacture of the fullerene by the combustion method, and it aims at offering the manufacture furnace of the fullerene which can manufacture fullerene cheaply and simple in large quantities.

[0006]

[Means for Solving the Problem] In the extensive and cheap manufacture approach of fullerene by the combustion method, this invention persons completed a header and this invention for the ability of the residence time of the fullerene precursor in combustion generation gas to be made uniform by controlling the condition of the flow of the combustion generation gas in the generation field of the fullerene in the manufacture furnace of fullerene, as a result of examining various optimal combustion conditions of the raw material in manufacture of fullerene. In the manufacture furnace of the fullerene concerning the 1st invention in alignment with said purpose, it is the manufacture furnace of fullerene which the feed hopper of a carbon content compound and the feed hopper of oxygen content gas are prepared in a manufacture furnace body, and this carbon content compound burns under this oxygen content gas, forms a combustion generation gas stream, and manufactures fullerene within this combustion generation gas stream, and average Reynolds number  $Re$  of said combustion generation gas stream is  $0 < Re \leq 2300$ .

[0007] It can prevent that self-circulating flow generates a carbon content compound in a combustion generation gas stream under oxygen content gas by controlling combustion and average Reynolds number  $Re$  of a combustion generation gas stream which was made to carry out a pyrolysis and was formed to  $0 < Re \leq 2300$ . If it can prevent that self-circulating flow occurs in a combustion generation gas stream, uneven migration of the fullerene precursor generated in the combustion generation gas stream is controlled, and all the residence times within the combustion generation gas stream of a fullerene precursor can be made uniform.

[0008] The manufacture furnace of the fullerene concerning the 2nd invention in alignment with said purpose The burner which has the feed hopper of a carbon content compound, and the feed hopper of oxygen content gas, and was formed in the manufacture furnace body, The 1st reaction band in which said carbon content compound is burned under said oxygen content gas, and a combustion gas style is formed, Supply the carbon content compound which blows in into said combustion gas style imported from this 1st reaction band succeeding said 1st reaction band, and mainly serves as a raw material from tubing, and a combustion generation gas stream is made to form. It is the manufacture furnace of the fullerene which has the 2nd reaction band by which fullerene is manufactured within this combustion generation gas stream, and average Reynolds number  $Re$  of said combustion generation gas stream in said 2nd reaction band is  $0 < Re \leq 2300$ .

[0009] It can prevent that the self-circulating flow of a combustion generation gas stream generates average Reynolds number  $Re$  of the combustion generation gas stream in the 2nd reaction band in the 2nd reaction band by controlling to  $0 < Re \leq 2300$ . If generating of the self-circulating flow of a combustion generation gas stream can be prevented, uneven migration of the fullerene precursor generated in the combustion generation gas stream is controlled, and the residence time in the 2nd reaction band of a fullerene precursor can be made uniform.

[0010] In the manufacture furnace of the fullerene concerning the 1st and 2nd invention, it is desirable that said average Reynolds number  $Re$  is  $0 < Re \leq 1500$ . By setting average Reynolds number  $Re$  of a

combustion generation gas stream to  $0 < Re \leq 1500$ , generating of the self-circulating flow within a combustion generation gas stream can be suppressed more. For this reason, the residence time within the combustion generation gas stream of the fullerene precursor formed within the combustion generation gas stream can be made more uniform.

[0011] In the manufacture furnace of the fullerene concerning the 1st and 2nd invention, it is still more desirable that said average Reynolds number  $Re$  is  $0 < Re \leq 1300$ . By setting average Reynolds number  $Re$  of a combustion generation gas stream to  $0 < Re \leq 1300$ , generating of the self-circulating flow within a combustion generation gas stream can be suppressed further. For this reason, the residence time within the combustion generation gas stream of the fullerene precursor formed within the combustion generation gas stream can be made still more uniform.

[0012] In the manufacture furnace of the fullerene concerning the 1st and 2nd invention, two or more feed hoppers of said carbon content compound and feed hoppers of said oxygen content gas are prepared every, respectively, and, as for each of these feed hoppers, it is desirable to become independent respectively, to separate distance and to carry out opening into said manufacture furnace body. It can become independent respectively, and the combustion condition within a manufacture furnace body can be made more into homogeneity from two or more feed hoppers which separated distance and were distributed by supplying a carbon content compound and oxygen content gas in a manufacture furnace body, respectively, consequently the temperature within a manufacture furnace body can be equalized.

[0013] As for the carbon content compound supplied from the feed hopper of said carbon content compound, in the manufacture furnace of the fullerene concerning the 1st and 2nd invention, it is desirable that the preheating is carried out. By raising the temperature of a carbon content compound, by raising to beyond the self-ignition temperature of for example, a carbon content compound, a carbon content compound can be burned to homogeneity and the temperature distribution within a manufacture furnace body can be made more into homogeneity.

[0014] As for said oxygen content gas, in the manufacture furnace of the fullerene concerning the 1st and 2nd invention, it is desirable to contain inert gas. Since the oxygen density in oxygen content gas is diluted by inert gas and falls, combustion within a manufacture furnace body can be changed into the condition that it was similar with the so-called elevated-temperature air combustion condition. For this reason, temperature within a manufacture furnace body can be made more into homogeneity and an elevated temperature. Here, oxygen gas and air can be used as a source of oxygen of oxygen content gas. Moreover, as inert gas, non-flammable gas, such as gaseous helium and argon gas, is used. In order to use non-flammable gas, excessive sub\*\* gas does not occur at the time of combustion, but it is desirable to generation of fullerene. Since sufficient heat energy to form the precursor of fullerene and for this grow to fullerene should just occur, there is especially no limit in the content rate of the inert gas in oxygen content gas. In addition, it is known that the yield of fullerene will become high, so that combustion flame temperature is high. Since addition of oxygen is effective as an approach of raising flame temperature, from a viewpoint of the yield of fullerene, oxygen gas is desirable as a source of oxygen.

[0015] In the manufacture furnace of the fullerene concerning the 1st and 2nd invention, it is desirable to make the pressure within said manufacture furnace body under into atmospheric pressure. It becomes possible by making the pressure within a manufacture furnace body under into atmospheric pressure, and rarefying the mixed state of a carbon content compound and oxygen content gas to make the condition that it was similar with the so-called elevated-temperature air combustion appear. For this reason, combustion advances to homogeneity and temperature within a manufacture furnace body can be made more into homogeneity and an elevated temperature.

[0016]

[Embodiment of the Invention] Then, referring to the attached drawing, it explains per gestalt of the operation which materialized this invention, and an understanding of this invention is presented. It is the explanatory view of the burner in which arrangement of the feed hopper of the whole manufacture furnace outline sectional view of the fullerene which the whole manufacture furnace outline sectional view of the fullerene which drawing 1 (A), and (B) require for the gestalt of operation of the 1st of this

invention, respectively, the explanatory view of the burner in which arrangement of the feed hopper of a carbon content compound and oxygen content gas was shown, drawing 2 (A), and (B) require for the gestalt of operation of the 2nd of this invention, respectively, a carbon content compound, and oxygen content gas be shown here. As shown in drawing 1 (A), the manufacture furnace 10 of the fullerene concerning the gestalt of operation of the 1st of this invention has the manufacture furnace body 11 and the burner 12 formed in the lower part of the manufacture furnace body 11. Hereafter, these are explained to a detail. The manufacture furnace body 11 is equipped with the cylindrical shape-like side-attachment-wall section 13 and the edge wall 15 which it connects with the end side of the side-attachment-wall section 13, and an outer diameter contracts gradually, and forms the exhaust port 14. The side-attachment-wall section 13 and the edge wall 15 consist of heat-resisting steel, such as stainless steel. Furthermore, refractories 16 are lined by the inner skin by the side of the other end of the side-attachment-wall section 13. As refractories 16, the refractory brick of the quality of an alumina and the unshaped refractories of the quality of an alumina can be used, for example. Moreover, the end side of the exhaust pipe which is not illustrated is connected to an exhaust port 14, and the other end side is connected to the exhaust air pump. For this reason, while changing the inside of the manufacture furnace body 11 into the reduced pressure condition of under atmospheric pressure, the combustion generation gas containing the fullerene generated within the manufacture furnace body 11 can be discharged outside from the inside of the manufacture furnace body 11.

[0017] The burner 12 attached in the other end side of the side-attachment-wall section 13 is equipped with the base 17 formed with heat-resisting steel, such as stainless steel, two or more feed hoppers 18 and 19 which are prepared in a base 17 and carry out the regurgitation of the carbon content compound, and two or more feed hoppers 20 and 21 which carry out the regurgitation of the oxygen content gas. As shown in drawing 1 (B), it distributes independently respectively and the feed hoppers 18 and 19 of a carbon content compound and the feed hoppers 20 and 21 of oxygen content gas are formed in the base 17. And opening of the one side of each feed hoppers 18, 19, 20, and 21 is carried out inside the manufacture furnace body 11, and the other side is connected to the supply pipe of the carbon content compound which is carrying out opening to the outside of the manufacture furnace body 11, and does not illustrate a gap, either, and the supply pipe of oxygen content gas, respectively. In addition, the heater is formed in the supply pipe of a carbon content compound, and the temperature of the carbon content compound at the time of being breathed out inside the manufacture furnace body 11 can be warmed to 200 degrees C. The configuration of each feed hoppers 18, 19, 20, and 21 which are carrying out opening inside the manufacture furnace body 11 may be arbitrary, and may have the shape of an indeterminate form, such as the shape of a polygon, such as the shape of an approximate circle form and an ellipse, a triangle, and a square, and a gourd mold. According to this invention persons' knowledge, in the configuration in which a circular twist also has a major axis and a minor axis like the diameter of an ellipse, or a rectangle, heating of oxygen content gas and the rate of dilution speed up more. Therefore, as feed hoppers 18 and 19 of a carbon content compound, the shape of an ellipse and an approximate circle form are desirable, as feed hoppers 20 and 21 of oxygen content gas, the shape of a rectangle, such as the shape of a slit, is desirable, and it is desirable especially to combine these.

[0018] If opening of the arrangement of the feed hoppers 18 and 19 of a carbon content compound and the feed hoppers 20 and 21 of oxygen content gas is respectively carried out inside the manufacture furnace body 11 independently, it will be possible with arbitration. Although various arrangement is employable according to the design condition of the manufacture furnace 10 of fullerene, such as a class of carbon content compound, and the number of each feed hoppers 18, 19, 20, and 21, if each feed hopper is arranged by turns on a concentric circle periphery to the axial center of the manufacture furnace 10 of fullerene in a hoop direction, since the combustion condition within the manufacture furnace body 11 will become more uniform for example, it is desirable. In this case, when the configuration of the feed hopper of oxygen content gas has a major axis and a minor axis, it is desirable to arrange so that the straight line prolonged from the major axis may pass along the core of a circle. Moreover, although you may project even if any feed hoppers 18, 19, 20, and 21 which are carrying out opening inside the manufacture furnace body 11 have the open end on the same flat surface substantially

with the front face of a base 17, the same flat-surface top is substantially good preferably. Each \*\* of the carbon content compound supplied in the manufacture furnace body 11 from the feed hoppers 18 and 19 of a carbon content compound and the feed hoppers 20 and 21 of oxygen content gas and oxygen content gas Although you may supply from the edge of each feed hoppers 18, 19, 20, and 21 at an angle of arbitration to the inside of the manufacture furnace body 11, so that it may become perpendicular substantially to a base 17 preferably further It is desirable to supply so that the carbon content compound and/or oxygen content gas which are supplied may be substantially spread in concentric circular from the core of the open end of feed hoppers 18, 19, 20, and 21.

[0019] Next, the manufacture approach of the fullerene which applied the manufacture furnace 10 of the fullerene concerning the gestalt of operation of the 1st of this invention is explained to a detail. From the feed hoppers 18 and 19 of a carbon content compound, oxygen content gas is supplied for the gas of the hydrocarbon which is a carbon content compound from the feed hoppers 20 and 21 of oxygen content gas, and a combustion gas style hot by burning these is generated toward the lower stream of a river of the manufacture furnace body 11. As oxygen content gas, the gas (for example, the concentration of inert gas can be adjusted in not more than 90 mol % exceeding 0 or 0) which mixed inert gas, such as argon gas, at a rate of arbitration can be used for the oxygen gas which is a source of oxygen. As a source of oxygen, from a viewpoint of the yield of fullerene, oxygen gas is desirable and air is desirable from a viewpoint of the ease of carrying out of acquisition of the source of oxygen etc. In order to raise especially combustion temperature, before these oxygen content gas is supplied in a furnace, it is desirable to become hot beforehand. As the approach of a preheating, what kind of well-known approaches, such as heat exchange with the combustion generation gas which used the heat exchanger, and the so-called regeneration burner, may be used. With [ the temperature of this preheating ] ordinary temperature [ beyond ], what kind of temperature is sufficient, but in order to gather the yield of fullerene, the high temperature is more desirable as much as possible. It is desirable more preferably that it is beyond the self-ignition temperature of a carbon content compound. As a carbon content compound, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as a carbon monoxide, natural gas, and petroleum gas, and a fuel oil, and creosote oil, can be used. Especially, it is desirable to use the aromatic series system hydrocarbon which refined these, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so desirable that its purity is close to 100% in case the purity of a raw material uses an aromatic series system hydrocarbon especially. Moreover, in order to gather the yield of fullerene, it is desirable to also dilute a carbon content compound using inert gas etc.

[0020] Then, the combustion generation gas stream in which it burns and pyrolyzes and a carbon content compound is formed under oxygen content gas is explained. While supplying in the manufacture furnace body 11 on the conditions on which the amount of the carbon content compound supplied from the feed hoppers 18 and 19 of a carbon content compound and the amount of oxygen gas supplied from the feed hoppers 20 and 21 of oxygen content gas are adjusted, and a carbon content compound combusts incompletely Combustion of a carbon content compound is started with an ignition means by which hold the inside of the manufacture furnace body 11, and an exhaust air pump does not illustrate it in the condition of 10 - 300torr more preferably under atmospheric pressure through the exhaust pipe which was connected to the exhaust port 14 and which is not illustrated. Here, a carbon content compound and oxygen content gas become independent respectively, and since it is breathed out in the manufacture furnace body 11 from the feed hoppers 18, 19, 20, and 21 of each plurality which separated distance and were distributed, they can make homogeneity the combustion condition within the manufacture furnace body 11. Moreover, it can promote that a carbon content compound burns in homogeneity by raising the temperature of a carbon content compound to 200 degrees C. furthermore, the thing which the oxygen gas concentration in oxygen content gas is diluted by inert gas, such as argon gas, and is being fallen -- in addition, since the pressure within the manufacture furnace body 11 has become under atmospheric pressure, the combustion condition within the manufacture furnace body 11 can be changed into the condition that it was similar with the elevated-temperature air combustion condition. Consequently, combustion of a carbon content compound advances to homogeneity, and can make temperature within

the manufacture furnace body 11 homogeneity and an elevated temperature (for example, 1000-1900 degrees C, preferably 1700-1900 degrees C).

[0021] 1000-1900 degrees C of temperature of the combustion zone within the manufacture furnace body 11 become a 1700-1900-degree C elevated temperature preferably by combustion of a carbon content compound, for example. For this reason, a non-burned carbon content compound is pyrolyzed easily, is evaporated, is diffused in the combustion gas which occurred by combustion of a carbon content compound, and forms combustion generation gas. That the pressure within the manufacture furnace body 11 is combustion in the thin condition that oxygen gas concentration is low, under in atmospheric pressure, and since homogeneity combustion is promoted and the temperature of combustion generation gas is uniform substantially in the direction perpendicular to the shaft orientations of the manufacture furnace body 11 further, it has been hard coming to generate self-circulating flow within the combustion generation gas stream formed by combustion generation gas. And since combustion generation gas is exhausted with the exhaust air pump from the exhaust port 14, the uniform flow by which a combustion generation gas stream goes to an exhaust port 14 from a burner 12 serves as a subject. Here, in the consistency of combustion generation gas, the mean velocity of  $5.3 \times 10^{-6} - 1.4 \times 10^{-4}$  kg/m/sec and combustion generation gas can estimate the viscosity of 0.01 - 0.006 kg/m<sup>3</sup> and combustion generation gas at 0.35-10m as range of the representation bore of 0.01 - 4 m/sec and the manufacture furnace body 11. Consequently, average Reynolds number Re of a combustion generation gas stream can be set to  $0 < Re \leq 2300$ .

[0022] By setting average Reynolds number Re to  $0 < Re \leq 2300$ , uneven migration of the fullerene precursor generated in the combustion generation gas stream is controlled, and the residence time within the combustion generation gas stream of a fullerene precursor can be made uniform. Consequently, it becomes possible to raise the yield of fullerene. Moreover, by controlling the representation bore and combustion condition of the manufacture furnace body 11, the consistency of combustion generation gas, the viscosity of combustion generation gas, and the mean velocity of combustion generation gas can be adjusted, respectively, and average Reynolds number Re of a combustion generation gas stream can be set to  $0 < Re \leq 1500$  and also  $0 < Re \leq 1300$ . The residence time within the combustion generation gas stream of the fullerene precursor which suppressed more generating of the self-circulating flow within a combustion generation gas stream, and was formed within the combustion generation gas stream can be made more uniform by setting average Reynolds number Re to  $0 < Re \leq 1500$  and also  $0 < Re \leq 1300$ . Consequently, it becomes possible to raise the yield of fullerene more.

[0023] As shown in drawing 2 (A), the manufacture furnace 22 of the fullerene concerning the gestalt of operation of the 2nd of this invention has the manufacture furnace body 23 and the burner 24 formed in the lower part of the manufacture furnace body 23. Hereafter, these are explained to a detail. The manufacture furnace body 23 is equipped with the cylindrical shape-like side-attachment-wall section 25 and the edge wall 27 which it connects with the end side of the side-attachment-wall section 25, and an outer diameter contracts gradually, and forms the exhaust port 26. The side-attachment-wall section 25 and the edge wall 27 consist of heat-resistant steel, such as stainless steel. Furthermore, the same refractories 16 as the gestalt of the 1st operation are lined by the inner skin by the side of the other end of the side-attachment-wall section 25. Moreover, the end side of the exhaust pipe which is not illustrated is connected to an exhaust port 26, and the other end side is connected to the exhaust air pump. For this reason, while changing the inside of the manufacture furnace body 23 into the reduced pressure condition of under atmospheric pressure, the combustion generation gas containing the fullerene generated within the manufacture furnace body 23 can be discharged outside from the inside of the manufacture furnace body 23. Furthermore, two or more entrainment tubing 28 and 29 which carries out the regurgitation of the carbon content compound which mainly serves as a raw material, respectively into the manufacture furnace body 23 is formed in the edge wall [ of the side-attachment-wall section 25 ] 27, and side-attachment-wall section 25 side of the edge wall 27. Opening is carried out inside the manufacture furnace body 23, the supply pipe with which the heater was formed and which is not illustrated is connected to the other end side, and the end side of the entrainment tubing 28 and 29 can warm the temperature of the carbon content compound which is blown into the interior of the

manufacture furnace body 23 and which mainly serves as a raw material to 200 degrees C. And the 1st reaction band consists of fields surrounded in the side-attachment-wall part to the location in which it blows in into from the burner 24 and other end side of the side-attachment-wall section 25, and tubing 28 is formed. Moreover, the 2nd reaction band consists of fields from the location in which the entrainment tubing 28 is formed to an exhaust port 26.

[0024] The burner 24 is equipped with the base 30 formed with heat-resisting steel, such as stainless steel, two or more feed hoppers 31 and 32 which carry out the regurgitation of the carbon content compound prepared in the base 30, and two or more feed hoppers 33 which carry out the regurgitation of the oxygen content gas. As shown in drawing 2 (B), it distributes independently respectively and the feed hoppers 31 and 32 of a carbon content compound and the feed hopper 33 of oxygen content gas are formed in the base 30. And opening of the one side of each feed hoppers 31, 32, and 33 is carried out inside the manufacture furnace body 23, and the other side is connected to the supply pipe of the carbon content compound which is carrying out opening to the outside of the manufacture furnace body 23, and does not illustrate a gap, either, and the supply pipe of oxygen content gas, respectively. In addition, the heater is formed in the supply pipe of a carbon content compound, and the temperature at the time of a carbon content compound being breathed out inside the manufacture furnace body 23 from the feed hoppers 31 and 32 of a carbon content compound can be warmed to 200 degrees C.

[0025] The configuration of each feed hoppers 31, 32, and 33 which are carrying out opening to the manufacture furnace body 23 inside may be arbitrary, and may have the shape of an indeterminate form, such as the shape of a polygon, such as the shape of an approximate circle form and an ellipse, a triangle, and a square, and a gourd mold. If opening of the arrangement of the feed hoppers 31 and 32 of a carbon content compound and the feed hopper 33 of oxygen content gas is respectively carried out inside the manufacture furnace body 23 independently, it is arbitrary. Although various arrangement is employable according to the design condition of the manufacture furnace 22 of fullerene, such as a class of carbon content compound, and the number of each feed hoppers of a carbon content compound and oxygen content gas, if each feed hopper is arranged on a concentric circle periphery to the axial center of the manufacture furnace 22 of fullerene, since the combustion condition within the manufacture furnace body 23 will become more uniform for example, it is desirable. Moreover, although you may project even if any feed hoppers 31, 32, and 33 which are carrying out opening inside the manufacture furnace body 23 have the open end on the same flat surface substantially with the front face of a base 30, the same flat-surface top is substantially good preferably. Each \*\* of the carbon content compound supplied in the manufacture furnace body 23 from the feed hoppers 31 and 32 of a carbon content compound and the feed hopper 33 of oxygen content gas and oxygen content gas Although you may supply from the edge of each feed hoppers 31, 32, and 33 at an angle of arbitration to the inside of the manufacture furnace body 23, so that it may become perpendicular substantially to a base 30 preferably further It is desirable to supply so that the carbon content compound and/or oxygen content gas which are supplied may be substantially spread in concentric circular from the core of the open end of feed hoppers 31, 32, and 33.

[0026] Next, the manufacture approach of the fullerene which applied the manufacture furnace 22 of the fullerene concerning the gestalt of operation of the 2nd of this invention is explained to a detail. From the feed hoppers 31 and 32 of a carbon content compound, oxygen content gas is supplied for hydrocarbon gas from the feed hopper 33 of oxygen content gas, and a combustion gas style hot by burning these is generated toward the lower stream of a river of the manufacture furnace body 23, for example. As oxygen content gas, the gas (for example, the concentration of inert gas can be adjusted in not more than 90 mol % exceeding 0 or 0) which mixed inert gas, such as argon gas, at a rate of arbitration can be used for the oxygen gas which is a source of oxygen. As a source of oxygen, from a viewpoint of the yield of fullerene, oxygen gas is desirable and air is desirable from a viewpoint of the ease of carrying out of acquisition of the source of oxygen etc. In order to raise especially combustion temperature, before these oxygen content gas is supplied in a furnace, it is desirable to become hot beforehand. As the approach of a preheating, what kind of well-known approaches, such as heat exchange with the combustion generation gas which used the heat exchanger, and the so-called

regeneration burner, may be used. With [ the temperature of this preheating ] ordinary temperature [ beyond ], what kind of temperature is sufficient, but in order to gather the yield of fullerene, the high temperature is more desirable as much as possible. It is desirable more preferably that it is beyond the self-ignition temperature of a carbon content compound.

[0027] Then, the combustion gas style in which the carbon content compound supplied from feed hoppers 31 and 32 is formed by burning, and the combustion generation gas stream in which the carbon content compound which was supplied from the entrainment tubing 28 and 29, and which mainly serves as a raw material pyrolyzes, and is formed in a combustion gas style are explained. In the 1st reaction band, the carbon content compound supplied to the interior of the manufacture furnace body 23 from feed hoppers 31 and 32 is burned by the oxygen content gas supplied from the feed hopper 33, and a hot combustion gas style is generated toward the lower stream of a river of the manufacture furnace body 23. In this 1st reaction band, it may be the purpose to generate hot combustion gas and that combustion method may be what kind of well-known combustion methods, such as premixed combustion, diffusive burning, laminar-flow combustion, turbulent flow combustion, and elevated-temperature air combustion. Combustion may be perfect combustion or may be incomplete combustion. However, the combustion by the lean mixture whose oxygen required for combustion is more than the amount of stoichiometry oxygen of the combustion in the 1st reaction band is preferably better. As oxygen content gas, the gas which mixed inert gas, such as argon gas, at a rate of arbitration can be used for the air which is a source of oxygen, and oxygen gas. Pure oxygen may be used in order to suppress generating of NO<sub>x</sub> especially in elevated-temperature combustion. In order to gather the yield of fullerene, it is desirable to dilute using inert gas in a combustion process. In addition, although it may be made to mix beforehand as oxygen content gas and you may supply, the carbon content compound supplied from feed hoppers 31 and 32 may be mixed, and inert gas may be supplied, even if it supplies from the exclusive nozzle for supply.

[0028] As a carbon content compound supplied from a burner 24, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as a carbon monoxide, natural gas, and petroleum gas, a fuel oil, benzene, and toluene, and creosote oil, can be used. Especially, hydrocarbon gas is desirable. Moreover, although what is necessary is for fullerene to obtain just to adjust suitably the mean temperature in the 1st reaction band at the time of fullerene manufacture, its 1600 degrees C or more 1300 degrees C or more are still more preferably good preferably. This is because the productivity of fullerene goes up, so that the temperature of combustion gas is an elevated temperature. In addition, if temperature becomes high too much, the productivity of fullerene may fall. Moreover, as for the furnace internal pressure within the manufacture furnace body 23, it is desirable that it is under atmospheric pressure, and the more desirable range is 10 - 300torr.

[0029] While making it an elevated temperature further by supplying the carbon content compound which blows in in the 2nd reaction band in the style of [ which was formed in the 1st reaction band ] combustion gas, and mainly serves as a raw material from tubing 28 and 29, and burning a part of this carbon content compound You pyrolyze the remaining carbon content compounds, you make it spread in a combustion gas style, a combustion generation gas stream is formed, and fullerene is made to generate within this combustion generation gas stream. In order to carry out partial combustion of the carbon content compound which mainly serves as a raw material, it is good also considering the combustion in the 1st reaction band as hyperoxia so that oxygen may remain. Moreover, the supply nozzle of oxygen content gas may be installed in the 2nd reaction band. Under the present circumstances, as for the carbon content compound which is supplied into combustion gas and which mainly serves as a raw material, it is desirable to be supplied in the manufacture furnace body 23 as much as possible at homogeneity. For this reason, it is desirable to be equally arranged so well that many by the number of the entrainment tubing 28 and 29 of the carbon content compound installed in the 2nd reaction band in a furnace. Moreover, it is more desirable for the cross-section configuration of \*\*\*\* not to change, although the configuration of the 2nd reaction band is also arbitrary. When the cross-section configuration of passage changes in the 1st reaction band and the 2nd reaction band, in case a combustion gas style imports the reason into the 2nd reaction band, it is for the yield of the

fullerene which turbulence, consequently the flow of the combustion generation gas by which the carbon content compound which mainly serves as a raw material is supplied and formed will also be confused, and flow generates to fall. When the cross-section configuration of the passage of combustion generation gas changes in the process which fullerene generates and turbulence of flow arises, it is for the yield of the fullerene to generate to fall.

[0030] Although what is necessary is just to choose the mean temperature of the 2nd reaction band suitably by the fullerene to manufacture, in order for the carbon content compound which mainly serves as a raw material to pyrolyze and to evaporate and react to homogeneity, it is desirable that it is an elevated temperature enough. It is desirable that it is specifically 1000 degrees C or more, and it is especially desirable that it is 1700-1900 degrees C 1000-1600 degrees C especially. Moreover, in the 2nd reaction band, it is desirable to control the oxygen density in combustion generation gas as much as possible. It is because the Lord of a reaction band, i.e., the 2nd reaction band, has the thing of the carbon content compound used as a raw material which combustion takes place actively in part, therefore the ununiformity of a reaction band produces when oxygen exists so much in combustion generation gas. the oxygen density in combustion generation gas -- desirable -- less than [ 3vol% ] -- it is 0.05 - 1vol% still more preferably. As a carbon content compound which mainly serves as a raw material, the thing of well-known arbitration can be used conventionally. For example, aromatic series system hydrocarbons, such as benzene, toluene, a xylene, naphthalene, and an anthracene, Coal system hydrocarbons, such as creosote oil and a carboxylic-acid oil, ethylene heavy-ends oil, Aliphatic saturated hydrocarbon, such as petroleum system heavy oil, such as FCC oil (fluidized-catalytic-cracking residue oil), acetylene series unsaturated hydrocarbon, the hydrocarbon of ethylene series, a pentane, and a hexane, etc. is mentioned, and these may be mixed and used at a rate of independent or arbitration. It is desirable to use the aromatic series system hydrocarbon refined especially, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of the carbon content compound which mainly serves as a raw material uses an aromatic series system hydrocarbon especially.

[0031] 1000-1900 degrees C of temperature of the 2nd reaction band are a 1700-1900-degree C elevated temperature preferably, for example. For this reason, the carbon content compound which was supplied into the combustion gas style imported from the 1st reaction band and which mainly serves as a raw material is pyrolyzed easily, is evaporated and diffused, and forms a combustion generation gas stream. the pressure within the manufacture furnace body 23 is combustion in the thin condition that oxygen gas concentration is low, under in atmospheric pressure -- further -- the temperature of combustion generation gas -- the shaft orientations of the manufacture furnace body 11 -- a perpendicular direction -- if -- since it is uniform substantially, it has been hard coming to generate self-circulating flow within the combustion generation gas stream formed by combustion generation gas And since combustion generation gas is exhausted with the exhaust air pump from the exhaust port 26, the uniform flow by which a combustion generation gas stream goes to an exhaust port 26 from a burner 24 serves as a subject. Here, in the consistency of combustion generation gas, the mean velocity of  $5.3 \times 10^{-6} - 1.4 \times 10^{-4}$  kg/m/sec and combustion generation gas can estimate the viscosity of 0.01 - 0.006 kg/m<sup>3</sup> and combustion generation gas at 0.35-10m as range of the representation bore of 0.01 - 4 m/sec and the manufacture furnace body 23. Consequently, average Reynolds number Re of a combustion generation gas stream can be set to  $0 < Re \leq 2300$ .

[0032] By setting average Reynolds number Re to  $0 < Re \leq 2300$ , uneven migration of the fullerene precursor generated in the combustion generation gas stream is controlled, and the residence time within the combustion generation gas stream of a fullerene precursor can be made uniform. Consequently, it becomes possible to raise the yield of fullerene. Moreover, by controlling the representation bore and combustion condition of the manufacture furnace body 23, the consistency of combustion generation gas, the viscosity of combustion generation gas, and the mean velocity of combustion generation gas can be adjusted, respectively, and average Reynolds number Re of a combustion generation gas stream can be set to  $0 < Re \leq 1500$  and also  $0 < Re \leq 1300$ . The residence time within the combustion generation gas stream of the fullerene precursor which suppressed more generating of the self-circulating flow within a

combustion generation gas stream, and was formed within the combustion generation gas stream can be made more uniform by setting average Reynolds number  $Re$  to  $0 < Re \leq 1500$  and also  $0 < Re \leq 1300$ . Consequently, it becomes possible to raise the yield of fullerene more.

[0033] As mentioned above, what is necessary is not to limit this invention to the gestalt of this operation, and just to choose the die length of the 2nd reaction band suitably according to the magnitude of a fission reactor, the class of fullerene to manufacture, etc., although the gestalt of operation of this invention was explained. Moreover, although the location of the 2nd reaction band was established in the upper part side of the 1st reaction band as shown in drawing 2 (A) Since the 2nd reaction band should just be prepared to the flow of the combustion gas style formed in the 1st reaction band succeeding the downstream Even if it forms in the outside of the first reaction band so that the 1st reaction band may be surrounded by controlling the flow direction of a combustion gas style, you may form inside the 1st reaction band so that it may be enclosed by the 1st reaction band. Furthermore, if it controls so that irregular flow, such as a turbulent flow, does not occur in combustion generation gas by supply of the carbon content compound which mainly serves as a raw material, the location of entrainment tubing can be set as arbitration.

[0034]

[Effect of the Invention] In the manufacture furnace of the fullerene according to claim 3 to 8 subordinate to claim 1 and this, since average Reynolds number  $Re$  of a combustion generation gas stream is  $0 < Re \leq 2300$ , equalization of the residence time of the fullerene precursor within a combustion generation gas stream can be attained, and it becomes possible to raise the yield of fullerene.

[0035] In the manufacture furnace of the fullerene according to claim 3 to 8 subordinate to claim 2 and this, since average Reynolds number  $Re$  of the combustion generation gas stream in the 2nd reaction band is  $0 < Re \leq 2300$ , equalization of the residence time of the fullerene precursor in the 2nd reaction band can be attained, and it becomes possible to raise the yield of fullerene.

[0036] Especially, since average Reynolds number  $Re$  is  $0 < Re \leq 1500$  in the manufacture furnace of fullerene according to claim 3, the residence time of the fullerene precursor formed within the combustion generation gas stream becomes more uniform, and it becomes possible to raise the yield of fullerene.

[0037] In the manufacture furnace of fullerene according to claim 4, since average Reynolds number  $Re$  is  $0 < Re \leq 1300$ , the residence time of the fullerene precursor within a combustion generation gas stream can be made still more uniform, and it becomes possible to raise the yield of fullerene further.

[0038] In the manufacture furnace of fullerene according to claim 5 Since two or more feed hoppers of a carbon content compound and feed hoppers of oxygen content gas are prepared every, respectively, each of these feed hoppers become independent respectively, distance is separated and opening is carried out into the manufacture furnace body By making the combustion condition within a manufacture furnace body equalize, it can prevent that make the temperature distribution within a manufacture furnace body into homogeneity, and self-circulating flow occurs in a combustion generation gas stream, and it becomes possible to generate fullerene efficiently.

[0039] In the manufacture furnace of fullerene according to claim 6, since the preheating of the carbon content compound supplied from the feed hopper of a carbon content compound is carried out, it can prevent that a carbon content compound burns in homogeneity, make the temperature distribution within a manufacture furnace body into homogeneity more, and self-circulating flow occurs in a combustion generation gas stream, and it becomes possible to generate fullerene more efficiently.

[0040] In the manufacture furnace of fullerene according to claim 7, since oxygen content gas contains inert gas, by making the temperature within a manufacture furnace body equalize more by burning by reducing an oxygen density, it can prevent that self-circulating flow occurs in a combustion generation gas stream, and it becomes possible to generate fullerene efficiently. Moreover, temperature within a manufacture furnace body is made more into an elevated temperature by burning by reducing an oxygen density, formation of a fullerene precursor can be promoted, and it becomes possible to raise the yield of fullerene more.

[0041] In the manufacture furnace of fullerene according to claim 8, since the pressure within a

manufacture furnace body is under atmospheric pressure, it can prevent that make the temperature within a manufacture furnace body equalize more by advancing a combustion reaction in the condition with thin carbon content compound and oxygen, and self-circulating flow occurs in a combustion generation gas stream, and it becomes possible to generate fullerene efficiently. Moreover, a carbon content compound and oxygen make temperature within a manufacture furnace body an elevated temperature more by advancing a combustion reaction in the thin condition, can promote formation of a fullerene precursor, and become possible [ raising the yield of fullerene more ]. furthermore, expensive gas like [ since an elevated temperature is acquired easily ] pure oxygen -- not using -- \*\*, for example, air, -- like -- acquisition -- it becomes generable [ fullerene ] using the easy source of oxygen, and it becomes possible to reduce the cost in fullerene manufacture sharply.

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**TECHNICAL FIELD**

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**[Field of the Invention]** This invention relates to the manufacture furnace of fullerene.

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PRIOR ART

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[Description of the Prior Art] Fullerene is the generic name of the third carbon allotrope which ranks second to a diamond and a graphite, and is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings as represented in C60, C70, etc. Although it is comparatively that existence of fullerene was finally checked and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.

- (1) Application to a superhard ingredient : since manufacture of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, use to an abrasion resistance material with added value is expected.
- (2) Application to drugs : research as an application of an anticancer agent, an acquired immunodeficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C60 derivative and an optical device.
- (3) Application to a superconducting material : if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.
- (4) Application to semi-conductor manufacture : it uses that resist structure is further strengthened with mixing C60 with a resist, and the application to next-generation semi-conductor manufacture is expected.

[0003] Also in the fullerene of various carbon numbers, C60 and C70 are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below as the manufacture approach of fullerene learned now is mentioned.

- (1) How to irradiate the pulse laser of a high energy consistency at the carbon target placed into laser vacuum deposition rare gas, evaporate a carbon atom, and compound fullerene. Into an electric furnace, the quartz tube with which inert gas flows is made to penetrate, and a graphite sample is placed into the quartz tube. If laser is irradiated from the upstream of the flow of gas at a graphite sample and a carbon atom is evaporated, the soot containing fullerene, such as C60 and C70, will adhere to the wall of the quartz tube near an electric furnace outlet. However, the evaporation per shot of a pulse laser is slight, and it is unsuitable for extensive manufacture of fullerene.
- (2) The approach to which carry out energization heating of the graphite rod in the gaseous helium ambient atmosphere below resistance heating method atmospheric pressure, and a carbon atom is made to sublimate. Since the electric resistance loss in the energization circuit constituted from a graphite rod is large, it is unsuitable for extensive manufacture of fullerene.
- (3) the-ten number of arc discharge methods -- the approach to which the carbon atom by the side of a lifting and an anode plate is made to sublimate arc discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium ambient atmosphere held to kPa, or having detached about 1-2mm. It is used for extensive manufacture of the fullerene in current and a works scale.
- (4) How to heat an eddy current to raw material graphite by radio frequency heating method high

frequency induction, heat raw material graphite with a sink and the Joule's heat to generate, and evaporate a carbon atom.

(5) The approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as combustion method helium, and oxygen. Manufacture effectiveness is not good at the point that several% of a benzene fuel serves as soot, and about 10% of them becomes fullerene. However, the soot which sub\*\* is observed as a method of mass-producing the fullerene which opposes an arc synthesis method in respect of a manufacturing installation being usable to liquid fuel etc., and simple etc.

(6) The approach of carrying out the pyrolysis of the naphthalene thermal decomposition method naphthalene at about 1000 degrees C.

[0004] As mentioned above, although the synthesis method of various fullerene by current is proposed, in which approach, the method of manufacturing fullerene in large quantities cheaply until now is not established. A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, and the manufacture approach of the fullerene by burning a carbon inclusion in a flame in the Patent Publication Heisei No. 507879 [ six to ] official report, and collecting condensates in it is indicated. When this manufacture approach burns a carbon inclusion in a flame, fullerene is manufactured and the fuel for combustion and the raw material of fullerene serve as the same carbon inclusion substantially. Although fullerene is contained in the soot-like matter and it is generated, a part of this soot-like matter is the so-called carbon black. As the manufacture approach of carbon black, the furnace method, a channel process, thermal \*\*, the acetylene method, etc. are learned, and the furnace method is mentioned as the industrial general manufacture approach. By this approach, use a cylinder-like fission reactor, supply oxygen content gas and fuels, such as air, to horizontal or a perpendicular direction to \*\*\* in the 1st reaction band, for example, and a combustion gas style is produced. Make it move to the 2nd reaction band which has the cross section which was installed in the lower stream of a river of furnace shaft orientations in the obtained combustion gas style, and was reduced as compared with the 1st reaction band, and supply coal-for-coke-making-ized hydrogen (stock oil), it is made then, to react into a combustion gas style, and carbon black is made to generate. Subsequently, the combustion gas style containing carbon black is moved to the 3rd reaction band on the lower stream of a river of the 2nd reaction band, cooling processing of spraying of cooling water etc. is performed in the style of combustion gas, combustion gas is quenched, a reaction is stopped, and carbon black is collected.

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## EFFECT OF THE INVENTION

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[Effect of the Invention] In the manufacture furnace of the fullerene according to claim 3 to 8 subordinate to claim 1 and this, since average Reynolds number Re of a combustion generation gas stream is  $0 < Re \leq 2300$ , equalization of the residence time of the fullerene precursor within a combustion generation gas stream can be attained, and it becomes possible to raise the yield of fullerene.

[0035] In the manufacture furnace of the fullerene according to claim 3 to 8 subordinate to claim 2 and this, since average Reynolds number Re of the combustion generation gas stream in the 2nd reaction band is  $0 < Re \leq 2300$ , equalization of the residence time of the fullerene precursor in the 2nd reaction band can be attained, and it becomes possible to raise the yield of fullerene.

[0036] Especially, since average Reynolds number Re is  $0 < Re \leq 1500$  in the manufacture furnace of fullerene according to claim 3, the residence time of the fullerene precursor formed within the combustion generation gas stream becomes more uniform, and it becomes possible to raise the yield of fullerene.

[0037] In the manufacture furnace of fullerene according to claim 4, since average Reynolds number Re is  $0 < Re \leq 1300$ , the residence time of the fullerene precursor within a combustion generation gas stream can be made still more uniform, and it becomes possible to raise the yield of fullerene further.

[0038] In the manufacture furnace of fullerene according to claim 5 Since two or more feed hoppers of a carbon content compound and feed hoppers of oxygen content gas are prepared every, respectively, each of these feed hoppers become independent respectively, distance is separated and opening is carried out into the manufacture furnace body By making the combustion condition within a manufacture furnace body equalize, it can prevent that make the temperature distribution within a manufacture furnace body into homogeneity, and self-circulating flow occurs in a combustion generation gas stream, and it becomes possible to generate fullerene efficiently.

[0039] In the manufacture furnace of fullerene according to claim 6, since the preheating of the carbon content compound supplied from the feed hopper of a carbon content compound is carried out, it can prevent that a carbon content compound burns in homogeneity, make the temperature distribution within a manufacture furnace body into homogeneity more, and self-circulating flow occurs in a combustion generation gas stream, and it becomes possible to generate fullerene more efficiently.

[0040] In the manufacture furnace of fullerene according to claim 7, since oxygen content gas contains inert gas, by making the temperature within a manufacture furnace body equalize more by burning by reducing an oxygen density, it can prevent that self-circulating flow occurs in a combustion generation gas stream, and it becomes possible to generate fullerene efficiently. Moreover, temperature within a manufacture furnace body is made more into an elevated temperature by burning by reducing an oxygen density, formation of a fullerene precursor can be promoted, and it becomes possible to raise the yield of fullerene more.

[0041] In the manufacture furnace of fullerene according to claim 8, since the pressure within a manufacture furnace body is under atmospheric pressure, it can prevent that make the temperature within a manufacture furnace body equalize more by advancing a combustion reaction in the condition with thin carbon content compound and oxygen, and self-circulating flow occurs in a combustion

generation gas stream, and it becomes possible to generate fullerene efficiently. Moreover, a carbon content compound and oxygen make temperature within a manufacture furnace body an elevated temperature more by advancing a combustion reaction in the thin condition, can promote formation of a fullerene precursor, and become possible [ raising the yield of fullerene more ]. furthermore, expensive gas like [ since an elevated temperature is acquired easily ] pure oxygen -- not using -- \*\*, for example, air, -- like -- acquisition -- it becomes generable [ fullerene ] using the easy source of oxygen, and it becomes possible to reduce the cost in fullerene manufacture sharply.

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## TECHNICAL PROBLEM

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[Problem(s) to be Solved by the Invention] However, by the manufacture approach of the above-mentioned usual carbon black, fullerene is hardly generated. Therefore, in manufacture of fullerene, it has been a big technical problem how the content rate of the fullerene contained in the soot-like matter obtained is raised. Usually, manufacture of fullerene is performed under reduced pressure and a diluent may be further introduced all over a reaction field. It is known whenever [ these reduced pressure ] that diluent concentration will affect the yield of fullerene. And when it is indicated that the yield of fullerene and a presentation change to the Patent Publication Heisei No. 507879 [ six to ] official report depending on the residence time in a flame and it manufactures fullerene with a combustion method, maintaining the residence time in a flame at homogeneity gathers the yield of fullerene, and it is considered to lead to carrying out a presentation to regularity. If a flame is made to form into the closed container generally, the rate of flow of the flame core where a combustion reaction is performed actively will be quick, and the rate of flow of the flame periphery section will become slow. For this reason, a back flow and contamination of the combustion generation gas from the upstream happen in the flame periphery section, and it becomes easy to generate self-circulation. Such self-circulation of combustion generation gas prevents local elevated-temperature-ization of flame temperature, and while it is effective in controlling generating of NO<sub>x</sub>, it also becomes the factor which brings about ununiformity-ization of the residence time of a fullerene precursor in the generation process of fullerene. That is, if self-circulation of combustion generation gas occurs, the residence time of the fullerene precursor which the residence time of the fullerene precursor which rode on this combustion generation gas stream through which it circulates in the phase which fullerene is generating in combustion generation gas became long, and did not ride on the combustion generation gas stream through which it circulates will become short. For this reason, the yield of fullerene falls in connection with the ununiformity of the residence time. Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and the residence time of such fullerene is controlled, and development of the technique whose manufacture is attained cheaply and simple in large quantities in fullerene is desired. This invention was made in view of this situation, and controls the condition of the flow of the combustion generation gas in the manufacture furnace of fullerene in manufacture of the fullerene by the combustion method, and it aims at offering the manufacture furnace of the fullerene which can manufacture fullerene cheaply and simple in large quantities.

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## MEANS

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[Means for Solving the Problem] In the extensive and cheap manufacture approach of fullerene by the combustion method, this invention persons completed a header and this invention for the ability of the residence time of the fullerene precursor in combustion generation gas to be made uniform by controlling the condition of the flow of the combustion generation gas in the generation field of the fullerene in the manufacture furnace of fullerene, as a result of examining various optimal combustion conditions of the raw material in manufacture of fullerene. In the manufacture furnace of the fullerene concerning the 1st invention in alignment with said purpose, it is the manufacture furnace of fullerene which the feed hopper of a carbon content compound and the feed hopper of oxygen content gas are prepared in a manufacture furnace body, and this carbon content compound burns under this oxygen content gas, forms a combustion generation gas stream, and manufactures fullerene within this combustion generation gas stream, and average Reynolds number Re of said combustion generation gas stream is  $0 < Re \leq 2300$ .

[0007] It can prevent that self-circulating flow generates a carbon content compound in a combustion generation gas stream under oxygen content gas by controlling combustion and average Reynolds number Re of a combustion generation gas stream which was made to carry out a pyrolysis and was formed to  $0 < Re \leq 2300$ . If it can prevent that self-circulating flow occurs in a combustion generation gas stream, uneven migration of the fullerene precursor generated in the combustion generation gas stream is controlled, and all the residence times within the combustion generation gas stream of a fullerene precursor can be made uniform.

[0008] The manufacture furnace of the fullerene concerning the 2nd invention in alignment with said purpose The burner which has the feed hopper of a carbon content compound, and the feed hopper of oxygen content gas, and was formed in the manufacture furnace body, The 1st reaction band in which said carbon content compound is burned under said oxygen content gas, and a combustion gas style is formed, Supply the carbon content compound which blows in into said combustion gas style imported from this 1st reaction band succeeding said 1st reaction band, and mainly serves as a raw material from tubing, and a combustion generation gas stream is made to form. It is the manufacture furnace of the fullerene which has the 2nd reaction band by which fullerene is manufactured within this combustion generation gas stream, and average Reynolds number Re of said combustion generation gas stream in said 2nd reaction band is  $0 < Re \leq 2300$ .

[0009] It can prevent that the self-circulating flow of a combustion generation gas stream generates average Reynolds number Re of the combustion generation gas stream in the 2nd reaction band in the 2nd reaction band by controlling to  $0 < Re \leq 2300$ . If generating of the self-circulating flow of a combustion generation gas stream can be prevented, uneven migration of the fullerene precursor generated in the combustion generation gas stream is controlled, and the residence time in the 2nd reaction band of a fullerene precursor can be made uniform.

[0010] In the manufacture furnace of the fullerene concerning the 1st and 2nd invention, it is desirable that said average Reynolds number Re is  $0 < Re \leq 1500$ . By setting average Reynolds number Re of a combustion generation gas stream to  $0 < Re \leq 1500$ , generating of the self-circulating flow within a

combustion generation gas stream can be suppressed more. For this reason, the residence time within the combustion generation gas stream of the fullerene precursor formed within the combustion generation gas stream can be made more uniform.

[0011] In the manufacture furnace of the fullerene concerning the 1st and 2nd invention, it is still more desirable that said average Reynolds number  $Re$  is  $0 < Re \leq 1300$ . By setting average Reynolds number  $Re$  of a combustion generation gas stream to  $0 < Re \leq 1300$ , generating of the self-circulating flow within a combustion generation gas stream can be suppressed further. For this reason, the residence time within the combustion generation gas stream of the fullerene precursor formed within the combustion generation gas stream can be made still more uniform.

[0012] In the manufacture furnace of the fullerene concerning the 1st and 2nd invention, two or more feed hoppers of said carbon content compound and feed hoppers of said oxygen content gas are prepared every, respectively, and, as for each of these feed hoppers, it is desirable to become independent respectively, to separate distance and to carry out opening into said manufacture furnace body. It can become independent respectively, and the combustion condition within a manufacture furnace body can be made more into homogeneity from two or more feed hoppers which separated distance and were distributed by supplying a carbon content compound and oxygen content gas in a manufacture furnace body, respectively, consequently the temperature within a manufacture furnace body can be equalized.

[0013] As for the carbon content compound supplied from the feed hopper of said carbon content compound, in the manufacture furnace of the fullerene concerning the 1st and 2nd invention, it is desirable that the preheating is carried out. By raising the temperature of a carbon content compound, by raising to beyond the self-ignition temperature of for example, a carbon content compound, a carbon content compound can be burned to homogeneity and the temperature distribution within a manufacture furnace body can be made more into homogeneity.

[0014] As for said oxygen content gas, in the manufacture furnace of the fullerene concerning the 1st and 2nd invention, it is desirable to contain inert gas. Since the oxygen density in oxygen content gas is diluted by inert gas and falls, combustion within a manufacture furnace body can be changed into the condition that it was similar with the so-called elevated-temperature air combustion condition. For this reason, temperature within a manufacture furnace body can be made more into homogeneity and an elevated temperature. Here, oxygen gas and air can be used as a source of oxygen of oxygen content gas. Moreover, as inert gas, non-flammable gas, such as gaseous helium and argon gas, is used. In order to use non-flammable gas, excessive sub\*\* gas does not occur at the time of combustion, but it is desirable to generation of fullerene. Since sufficient heat energy to form the precursor of fullerene and for this grow to fullerene should just occur, there is especially no limit in the content rate of the inert gas in oxygen content gas. In addition, it is known that the yield of fullerene will become high, so that combustion flame temperature is high. Since addition of oxygen is effective as an approach of raising flame temperature, from a viewpoint of the yield of fullerene, oxygen gas is desirable as a source of oxygen.

[0015] In the manufacture furnace of the fullerene concerning the 1st and 2nd invention, it is desirable to make the pressure within said manufacture furnace body under into atmospheric pressure. It becomes possible by making the pressure within a manufacture furnace body under into atmospheric pressure, and rarefying the mixed state of a carbon content compound and oxygen content gas to make the condition that it was similar with the so-called elevated-temperature air combustion appear. For this reason, combustion advances to homogeneity and temperature within a manufacture furnace body can be made more into homogeneity and an elevated temperature.

[0016]

[Embodiment of the Invention] Then, referring to the attached drawing, it explains per gestalt of the operation which materialized this invention, and an understanding of this invention is presented. It is the explanatory view of the burner in which arrangement of the feed hopper of the whole manufacture furnace outline sectional view of the fullerene which the whole manufacture furnace outline sectional view of the fullerene which drawing 1 (A), and (B) require for the gestalt of operation of the 1st of this invention, respectively, the explanatory view of the burner in which arrangement of the feed hopper of a

carbon content compound and oxygen content gas was shown, drawing 2 (A), and (B) require for the gestalt of operation of the 2nd of this invention, respectively, a carbon content compound, and oxygen content gas be shown here. As shown in drawing 1 (A), the manufacture furnace 10 of the fullerene concerning the gestalt of operation of the 1st of this invention has the manufacture furnace body 11 and the burner 12 formed in the lower part of the manufacture furnace body 11. Hereafter, these are explained to a detail. The manufacture furnace body 11 is equipped with the cylindrical shape-like side-attachment-wall section 13 and the edge wall 15 which it connects with the end side of the side-attachment-wall section 13, and an outer diameter contracts gradually, and forms the exhaust port 14. The side-attachment-wall section 13 and the edge wall 15 consist of heat-resisting steel, such as stainless steel. Furthermore, refractories 16 are lined by the inner skin by the side of the other end of the side-attachment-wall section 13. As refractories 16, the refractory brick of the quality of an alumina and the unshaped refractories of the quality of an alumina can be used, for example. Moreover, the end side of the exhaust pipe which is not illustrated is connected to an exhaust port 14, and the other end side is connected to the exhaust air pump. For this reason, while changing the inside of the manufacture furnace body 11 into the reduced pressure condition of under atmospheric pressure, the combustion generation gas containing the fullerene generated within the manufacture furnace body 11 can be discharged outside from the inside of the manufacture furnace body 11.

[0017] The burner 12 attached in the other end side of the side-attachment-wall section 13 is equipped with the base 17 formed with heat-resisting steel, such as stainless steel, two or more feed hoppers 18 and 19 which are prepared in a base 17 and carry out the regurgitation of the carbon content compound, and two or more feed hoppers 20 and 21 which carry out the regurgitation of the oxygen content gas. As shown in drawing 1 (B), it distributes independently respectively and the feed hoppers 18 and 19 of a carbon content compound and the feed hoppers 20 and 21 of oxygen content gas are formed in the base 17. And opening of the one side of each feed hoppers 18, 19, 20, and 21 is carried out inside the manufacture furnace body 11, and the other side is connected to the supply pipe of the carbon content compound which is carrying out opening to the outside of the manufacture furnace body 11, and does not illustrate a gap, either, and the supply pipe of oxygen content gas, respectively. In addition, the heater is formed in the supply pipe of a carbon content compound, and the temperature of the carbon content compound at the time of being breathed out inside the manufacture furnace body 11 can be warmed to 200 degrees C. The configuration of each feed hoppers 18, 19, 20, and 21 which are carrying out opening inside the manufacture furnace body 11 may be arbitrary, and may have the shape of an indeterminate form, such as the shape of a polygon, such as the shape of an approximate circle form and an ellipse, a triangle, and a square, and a gourd mold. According to this invention persons' knowledge, in the configuration in which a circular twist also has a major axis and a minor axis like the diameter of an ellipse, or a rectangle, heating of oxygen content gas and the rate of dilution speed up more. Therefore, as feed hoppers 18 and 19 of a carbon content compound, the shape of an ellipse and an approximate circle form are desirable, as feed hoppers 20 and 21 of oxygen content gas, the shape of a rectangle, such as the shape of a slit, is desirable, and it is desirable especially to combine these.

[0018] If opening of the arrangement of the feed hoppers 18 and 19 of a carbon content compound and the feed hoppers 20 and 21 of oxygen content gas is respectively carried out inside the manufacture furnace body 11 independently, it will be possible with arbitration. Although various arrangement is employable according to the design condition of the manufacture furnace 10 of fullerene, such as a class of carbon content compound, and the number of each feed hoppers 18, 19, 20, and 21, if each feed hopper is arranged by turns on a concentric circle periphery to the axial center of the manufacture furnace 10 of fullerene in a hoop direction, since the combustion condition within the manufacture furnace body 11 will become more uniform for example, it is desirable. In this case, when the configuration of the feed hopper of oxygen content gas has a major axis and a minor axis, it is desirable to arrange so that the straight line prolonged from the major axis may pass along the core of a circle. Moreover, although you may project even if any feed hoppers 18, 19, 20, and 21 which are carrying out opening inside the manufacture furnace body 11 have the open end on the same flat surface substantially with the front face of a base 17, the same flat-surface top is substantially good preferably. Each \*\* of the

carbon content compound supplied in the manufacture furnace body 11 from the feed hoppers 18 and 19 of a carbon content compound and the feed hoppers 20 and 21 of oxygen content gas and oxygen content gas. Although you may supply from the edge of each feed hoppers 18, 19, 20, and 21 at an angle of arbitration to the inside of the manufacture furnace body 11, so that it may become perpendicular substantially to a base 17 preferably further. It is desirable to supply so that the carbon content compound and/or oxygen content gas which are supplied may be substantially spread in concentric circular from the core of the open end of feed hoppers 18, 19, 20, and 21.

[0019] Next, the manufacture approach of the fullerene which applied the manufacture furnace 10 of the fullerene concerning the gestalt of operation of the 1st of this invention is explained to a detail. From the feed hoppers 18 and 19 of a carbon content compound, oxygen content gas is supplied for the gas of the hydrocarbon which is a carbon content compound from the feed hoppers 20 and 21 of oxygen content gas, and a combustion gas style hot by burning these is generated toward the lower stream of a river of the manufacture furnace body 11. As oxygen content gas, the gas (for example, the concentration of inert gas can be adjusted in not more than 90 mol % exceeding 0 or 0) which mixed inert gas, such as argon gas, at a rate of arbitration can be used for the oxygen gas which is a source of oxygen. As a source of oxygen, from a viewpoint of the yield of fullerene, oxygen gas is desirable and air is desirable from a viewpoint of the ease of carrying out of acquisition of the source of oxygen etc. In order to raise especially combustion temperature, before these oxygen content gas is supplied in a furnace, it is desirable to become hot beforehand. As the approach of a preheating, what kind of well-known approaches, such as heat exchange with the combustion generation gas which used the heat exchanger, and the so-called regeneration burner, may be used. With [ the temperature of this preheating ] ordinary temperature [ beyond ], what kind of temperature is sufficient, but in order to gather the yield of fullerene, the high temperature is more desirable as much as possible. It is desirable more preferably that it is beyond the self-ignition temperature of a carbon content compound. As a carbon content compound, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as a carbon monoxide, natural gas, and petroleum gas, and a fuel oil, and creosote oil, can be used. Especially, it is desirable to use the aromatic series system hydrocarbon which refined these, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so desirable that its purity is close to 100% in case the purity of a raw material uses an aromatic series system hydrocarbon especially. Moreover, in order to gather the yield of fullerene, it is desirable to also dilute a carbon content compound using inert gas etc.

[0020] Then, the combustion generation gas stream in which it burns and pyrolyzes and a carbon content compound is formed under oxygen content gas is explained. While supplying in the manufacture furnace body 11 on the conditions on which the amount of the carbon content compound supplied from the feed hoppers 18 and 19 of a carbon content compound and the amount of oxygen gas supplied from the feed hoppers 20 and 21 of oxygen content gas are adjusted, and a carbon content compound combusts incompletely. Combustion of a carbon content compound is started with an ignition means by which hold the inside of the manufacture furnace body 11, and an exhaust air pump does not illustrate it in the condition of 10 - 300 torr more preferably under atmospheric pressure through the exhaust pipe which was connected to the exhaust port 14 and which is not illustrated. Here, a carbon content compound and oxygen content gas become independent respectively, and since it is breathed out in the manufacture furnace body 11 from the feed hoppers 18, 19, 20, and 21 of each plurality which separated distance and were distributed, they can make homogeneity the combustion condition within the manufacture furnace body 11. Moreover, it can promote that a carbon content compound burns in homogeneity by raising the temperature of a carbon content compound to 200 degrees C. furthermore, the thing which the oxygen gas concentration in oxygen content gas is diluted by inert gas, such as argon gas, and is being fallen -- in addition, since the pressure within the manufacture furnace body 11 has become under atmospheric pressure, the combustion condition within the manufacture furnace body 11 can be changed into the condition that it was similar with the elevated-temperature air combustion condition. Consequently, combustion of a carbon content compound advances to homogeneity, and can make temperature within the manufacture furnace body 11 homogeneity and an elevated temperature (for example, 1000-1900

degrees C, preferably 1700-1900 degrees C).

[0021] 1000-1900 degrees C of temperature of the combustion zone within the manufacture furnace body 11 become a 1700-1900-degree C elevated temperature preferably by combustion of a carbon content compound, for example. For this reason, a non-burned carbon content compound is pyrolyzed easily, is evaporated, is diffused in the combustion gas which occurred by combustion of a carbon content compound, and forms combustion generation gas. That the pressure within the manufacture furnace body 11 is combustion in the thin condition that oxygen gas concentration is low, under in atmospheric pressure, and since homogeneity combustion is promoted and the temperature of combustion generation gas is uniform substantially in the direction perpendicular to the shaft orientations of the manufacture furnace body 11 further, it has been hard coming to generate self-circulating flow within the combustion generation gas stream formed by combustion generation gas. And since combustion generation gas is exhausted with the exhaust air pump from the exhaust port 14, the uniform flow by which a combustion generation gas stream goes to an exhaust port 14 from a burner 12 serves as a subject. Here, in the consistency of combustion generation gas, the mean velocity of  $5.3 \times 10^{-6} - 1.4 \times 10^{-4}$  kg/m/sec and combustion generation gas can estimate the viscosity of 0.01 - 0.006 kg/m<sup>3</sup> and combustion generation gas at 0.35-10m as range of the representation bore of 0.01 - 4 m/sec and the manufacture furnace body 11. Consequently, average Reynolds number Re of a combustion generation gas stream can be set to  $0 < Re \leq 2300$ .

[0022] By setting average Reynolds number Re to  $0 < Re \leq 2300$ , uneven migration of the fullerene precursor generated in the combustion generation gas stream is controlled, and the residence time within the combustion generation gas stream of a fullerene precursor can be made uniform. Consequently, it becomes possible to raise the yield of fullerene. Moreover, by controlling the representation bore and combustion condition of the manufacture furnace body 11, the consistency of combustion generation gas, the viscosity of combustion generation gas, and the mean velocity of combustion generation gas can be adjusted, respectively, and average Reynolds number Re of a combustion generation gas stream can be set to  $0 < Re \leq 1500$  and also  $0 < Re \leq 1300$ . The residence time within the combustion generation gas stream of the fullerene precursor which suppressed more generating of the self-circulating flow within a combustion generation gas stream, and was formed within the combustion generation gas stream can be made more uniform by setting average Reynolds number Re to  $0 < Re \leq 1500$  and also  $0 < Re \leq 1300$ . Consequently, it becomes possible to raise the yield of fullerene more.

[0023] As shown in drawing 2 (A), the manufacture furnace 22 of the fullerene concerning the gestalt of operation of the 2nd of this invention has the manufacture furnace body 23 and the burner 24 formed in the lower part of the manufacture furnace body 23. Hereafter, these are explained to a detail. The manufacture furnace body 23 is equipped with the cylindrical shape-like side-attachment-wall section 25 and the edge wall 27 which it connects with the end side of the side-attachment-wall section 25, and an outer diameter contracts gradually, and forms the exhaust port 26. The side-attachment-wall section 25 and the edge wall 27 consist of heat-resisting steel, such as stainless steel. Furthermore, the same refractories 16 as the gestalt of the 1st operation are lined by the inner skin by the side of the other end of the side-attachment-wall section 25. Moreover, the end side of the exhaust pipe which is not illustrated is connected to an exhaust port 26, and the other end side is connected to the exhaust air pump. For this reason, while changing the inside of the manufacture furnace body 23 into the reduced pressure condition of under atmospheric pressure, the combustion generation gas containing the fullerene generated within the manufacture furnace body 23 can be discharged outside from the inside of the manufacture furnace body 23. Furthermore, two or more entrainment tubing 28 and 29 which carries out the regurgitation of the carbon content compound which mainly serves as a raw material, respectively into the manufacture furnace body 23 is formed in the edge wall [ of the side-attachment-wall section 25 ] 27, and side-attachment-wall section 25 side of the edge wall 27. Opening is carried out inside the manufacture furnace body 23, the supply pipe with which the heater was formed and which is not illustrated is connected to the other end side, and the end side of the entrainment tubing 28 and 29 can warm the temperature of the carbon content compound which is blown into the interior of the manufacture furnace body 23 and which mainly serves as a raw material to 200 degrees C. And the 1st

reaction band consists of fields surrounded in the side-attachment-wall part to the location in which it blows in into from the burner 24 and other end side of the side-attachment-wall section 25, and tubing 28 is formed. Moreover, the 2nd reaction band consists of fields from the location in which the entrainment tubing 28 is formed to an exhaust port 26.

[0024] The burner 24 is equipped with the base 30 formed with heat-resisting steel, such as stainless steel, two or more feed hoppers 31 and 32 which carry out the regurgitation of the carbon content compound prepared in the base 30, and two or more feed hoppers 33 which carry out the regurgitation of the oxygen content gas. As shown in drawing 2 (B), it distributes independently respectively and the feed hoppers 31 and 32 of a carbon content compound and the feed hopper 33 of oxygen content gas are formed in the base 30. And opening of the one side of each feed hoppers 31, 32, and 33 is carried out inside the manufacture furnace body 23, and the other side is connected to the supply pipe of the carbon content compound which is carrying out opening to the outside of the manufacture furnace body 23, and does not illustrate a gap, either, and the supply pipe of oxygen content gas, respectively. In addition, the heater is formed in the supply pipe of a carbon content compound, and the temperature at the time of a carbon content compound being breathed out inside the manufacture furnace body 23 from the feed hoppers 31 and 32 of a carbon content compound can be warmed to 200 degrees C.

[0025] The configuration of each feed hoppers 31, 32, and 33 which are carrying out opening to the manufacture furnace body 23 inside may be arbitrary, and may have the shape of an indeterminate form, such as the shape of a polygon, such as the shape of an approximate circle form and an ellipse, a triangle, and a square, and a gourd mold. If opening of the arrangement of the feed hoppers 31 and 32 of a carbon content compound and the feed hopper 33 of oxygen content gas is respectively carried out inside the manufacture furnace body 23 independently, it is arbitrary. Although various arrangement is employable according to the design condition of the manufacture furnace 22 of fullerene, such as a class of carbon content compound, and the number of each feed hoppers of a carbon content compound and oxygen content gas, if each feed hopper is arranged on a concentric circle periphery to the axial center of the manufacture furnace 22 of fullerene, since the combustion condition within the manufacture furnace body 23 will become more uniform for example, it is desirable. Moreover, although you may project even if any feed hoppers 31, 32, and 33 which are carrying out opening inside the manufacture furnace body 23 have the open end on the same flat surface substantially with the front face of a base 30, the same flat-surface top is substantially good preferably. Each \*\* of the carbon content compound supplied in the manufacture furnace body 23 from the feed hoppers 31 and 32 of a carbon content compound and the feed hopper 33 of oxygen content gas and oxygen content gas Although you may supply from the edge of each feed hoppers 31, 32, and 33 at an angle of arbitration to the inside of the manufacture furnace body 23, so that it may become perpendicular substantially to a base 30 preferably further It is desirable to supply so that the carbon content compound and/or oxygen content gas which are supplied may be substantially spread in concentric circular from the core of the open end of feed hoppers 31, 32, and 33.

[0026] Next, the manufacture approach of the fullerene which applied the manufacture furnace 22 of the fullerene concerning the gestalt of operation of the 2nd of this invention is explained to a detail. From the feed hoppers 31 and 32 of a carbon content compound, oxygen content gas is supplied for hydrocarbon gas from the feed hopper 33 of oxygen content gas, and a combustion gas style hot by burning these is generated toward the lower stream of a river of the manufacture furnace body 23, for example. As oxygen content gas, the gas (for example, the concentration of inert gas can be adjusted in not more than 90 mol % exceeding 0 or 0) which mixed inert gas, such as argon gas, at a rate of arbitration can be used for the oxygen gas which is a source of oxygen. As a source of oxygen, from a viewpoint of the yield of fullerene, oxygen gas is desirable and air is desirable from a viewpoint of the ease of carrying out of acquisition of the source of oxygen etc. In order to raise especially combustion temperature, before these oxygen content gas is supplied in a furnace, it is desirable to become hot beforehand. As the approach of a preheating, what kind of well-known approaches, such as heat exchange with the combustion generation gas which used the heat exchanger, and the so-called regeneration burner, may be used. With [ the temperature of this preheating ] ordinary temperature

[ beyond ], what kind of temperature is sufficient, but in order to gather the yield of fullerene, the high temperature is more desirable as much as possible. It is desirable more preferably that it is beyond the self-ignition temperature of a carbon content compound.

[0027] Then, the combustion gas style in which the carbon content compound supplied from feed hoppers 31 and 32 is formed by burning, and the combustion generation gas stream in which the carbon content compound which was supplied from the entrainment tubing 28 and 29, and which mainly serves as a raw material pyrolyzes, and is formed in a combustion gas style are explained. In the 1st reaction band, the carbon content compound supplied to the interior of the manufacture furnace body 23 from feed hoppers 31 and 32 is burned by the oxygen content gas supplied from the feed hopper 33, and a hot combustion gas style is generated toward the lower stream of a river of the manufacture furnace body 23. In this 1st reaction band, it may be the purpose to generate hot combustion gas and that combustion method may be what kind of well-known combustion methods, such as premixed combustion, diffusive burning, laminar-flow combustion, turbulent flow combustion, and elevated-temperature air combustion. Combustion may be perfect combustion or may be incomplete combustion. However, the combustion by the lean mixture whose oxygen required for combustion is more than the amount of stoichiometry oxygen of the combustion in the 1st reaction band is preferably better. As oxygen content gas, the gas which mixed inert gas, such as argon gas, at a rate of arbitration can be used for the air which is a source of oxygen, and oxygen gas. Pure oxygen may be used in order to suppress generating of NOx especially in elevated-temperature combustion. In order to gather the yield of fullerene, it is desirable to dilute using inert gas in a combustion process. In addition, although it may be made to mix beforehand as oxygen content gas and you may supply, the carbon content compound supplied from feed hoppers 31 and 32 may be mixed, and inert gas may be supplied, even if it supplies from the exclusive nozzle for supply.

[0028] As a carbon content compound supplied from a burner 24, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as a carbon monoxide, natural gas, and petroleum gas, a fuel oil, benzene, and toluene, and creosote oil, can be used. Especially, hydrocarbon gas is desirable. Moreover, although what is necessary is for fullerene to obtain just to adjust suitably the mean temperature in the 1st reaction band at the time of fullerene manufacture, its 1600 degrees C or more 1300 degrees C or more are still more preferably good preferably. This is because the productivity of fullerene goes up, so that the temperature of combustion gas is an elevated temperature. In addition, if temperature becomes high too much, the productivity of fullerene may fall. Moreover, as for the furnace internal pressure within the manufacture furnace body 23, it is desirable that it is under atmospheric pressure, and the more desirable range is 10 - 300torr.

[0029] While making it an elevated temperature further by supplying the carbon content compound which blows in in the 2nd reaction band in the style of [ which was formed in the 1st reaction band ] combustion gas, and mainly serves as a raw material from tubing 28 and 29, and burning a part of this carbon content compound You pyrolyze the remaining carbon content compounds, you make it spread in a combustion gas style, a combustion generation gas stream is formed, and fullerene is made to generate within this combustion generation gas stream. In order to carry out partial combustion of the carbon content compound which mainly serves as a raw material, it is good also considering the combustion in the 1st reaction band as hyperoxia so that oxygen may remain. Moreover, the supply nozzle of oxygen content gas may be installed in the 2nd reaction band. Under the present circumstances, as for the carbon content compound which is supplied into combustion gas and which mainly serves as a raw material, it is desirable to be supplied in the manufacture furnace body 23 as much as possible at homogeneity. For this reason, it is desirable to be equally arranged so well that many by the number of the entrainment tubing 28 and 29 of the carbon content compound installed in the 2nd reaction band in a furnace. Moreover, it is more desirable for the cross-section configuration of \*\*\*\* not to change, although the configuration of the 2nd reaction band is also arbitrary. When the cross-section configuration of passage changes in the 1st reaction band and the 2nd reaction band, in case a combustion gas style imports the reason into the 2nd reaction band, it is for the yield of the fullerene which turbulence, consequently the flow of the combustion generation gas by which the carbon

content compound which mainly serves as a raw material is supplied and formed will also be confused, and flow generates to fall. When the cross-section configuration of the passage of combustion generation gas changes in the process which fullerene generates and turbulence of flow arises, it is for the yield of the fullerene to generate to fall.

[0030] Although what is necessary is just to choose the mean temperature of the 2nd reaction band suitably by the fullerene to manufacture, in order for the carbon content compound which mainly serves as a raw material to pyrolyze and to evaporate and react to homogeneity, it is desirable that it is an elevated temperature enough. It is desirable that it is specifically 1000 degrees C or more, and it is especially desirable that it is 1700-1900 degrees C 1000-1600 degrees C especially. Moreover, in the 2nd reaction band, it is desirable to control the oxygen density in combustion generation gas as much as possible. It is because the Lord of a reaction band, i.e., the 2nd reaction band, has the thing of the carbon content compound used as a raw material which combustion takes place actively in part, therefore the ununiformity of a reaction band produces when oxygen exists so much in combustion generation gas. the oxygen density in combustion generation gas -- desirable -- less than [ 3vol% ] -- it is 0.05 - 1vol% still more preferably. As a carbon content compound which mainly serves as a raw material, the thing of well-known arbitration can be used conventionally. For example, aromatic series system hydrocarbons, such as benzene, toluene, a xylene, naphthalene, and an anthracene, Coal system hydrocarbons, such as creosote oil and a carboxylic-acid oil, ethylene heavy-ends oil, Aliphatic saturated hydrocarbon, such as petroleum system heavy oil, such as FCC oil (fluidized-catalytic-cracking residue oil), acetylene series unsaturated hydrocarbon, the hydrocarbon of ethylene series, a pentane, and a hexane, etc. is mentioned, and these may be mixed and used at a rate of independent or arbitration. It is desirable to use the aromatic series system hydrocarbon refined especially, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of the carbon content compound which mainly serves as a raw material uses an aromatic series system hydrocarbon especially.

[0031] 1000-1900 degrees C of temperature of the 2nd reaction band are a 1700-1900-degree C elevated temperature preferably, for example. For this reason, the carbon content compound which was supplied into the combustion gas style imported from the 1st reaction band and which mainly serves as a raw material is pyrolyzed easily, is evaporated and diffused, and forms a combustion generation gas stream. the pressure within the manufacture furnace body 23 is combustion in the thin condition that oxygen gas concentration is low, under in atmospheric pressure -- further -- the temperature of combustion generation gas -- the shaft orientations of the manufacture furnace body 11 -- a perpendicular direction -- if -- since it is uniform substantially, it has been hard coming to generate self-circulating flow within the combustion generation gas stream formed by combustion generation gas And since combustion generation gas is exhausted with the exhaust air pump from the exhaust port 26, the uniform flow by which a combustion generation gas stream goes to an exhaust port 26 from a burner 24 serves as a subject. Here, in the consistency of combustion generation gas, the mean velocity of  $5.3 \times 10^{-6} - 1.4 \times 10^{-4}$  kg/m/sec and combustion generation gas can estimate the viscosity of  $0.01 - 0.006$  kg/m<sup>3</sup> and combustion generation gas at 0.35-10m as range of the representation bore of 0.01 - 4 m/sec and the manufacture furnace body 23. Consequently, average Reynolds number Re of a combustion generation gas stream can be set to  $0 < Re <= 2300$ .

[0032] By setting average Reynolds number Re to  $0 < Re <= 2300$ , uneven migration of the fullerene precursor generated in the combustion generation gas stream is controlled, and the residence time within the combustion generation gas stream of a fullerene precursor can be made uniform. Consequently, it becomes possible to raise the yield of fullerene. Moreover, by controlling the representation bore and combustion condition of the manufacture furnace body 23, the consistency of combustion generation gas, the viscosity of combustion generation gas, and the mean velocity of combustion generation gas can be adjusted, respectively, and average Reynolds number Re of a combustion generation gas stream can be set to  $0 < Re <= 1500$  and also  $0 < Re <= 1300$ . The residence time within the combustion generation gas stream of the fullerene precursor which suppressed more generating of the self-circulating flow within a combustion generation gas stream, and was formed within the combustion generation gas stream can be

made more uniform by setting average Reynolds number  $Re$  to  $0 < Re \leq 1500$  and also  $0 < Re \leq 1300$ . Consequently, it becomes possible to raise the yield of fullerene more.

[0033] As mentioned above, what is necessary is not to limit this invention to the gestalt of this operation, and just to choose the die length of the 2nd reaction band suitably according to the magnitude of a fission reactor, the class of fullerene to manufacture, etc., although the gestalt of operation of this invention was explained. Moreover, although the location of the 2nd reaction band was established in the upper part side of the 1st reaction band as shown in drawing 2 (A) Since the 2nd reaction band should just be prepared to the flow of the combustion gas style formed in the 1st reaction band succeeding the downstream Even if it forms in the outside of the first reaction band so that the 1st reaction band may be surrounded by controlling the flow direction of a combustion gas style, you may form inside the 1st reaction band so that it may be enclosed by the 1st reaction band. Furthermore, if it controls so that irregular flow, such as a turbulent flow, does not occur in combustion generation gas by supply of the carbon content compound which mainly serves as a raw material, the location of entrainment tubing can be set as arbitration.

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[Translation done.]

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## DESCRIPTION OF DRAWINGS

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### [Brief Description of the Drawings]

[Drawing 1] (A) and (B) are the whole manufacture furnace outline sectional view of the fullerene concerning the gestalt of operation of the 1st of this invention, and the explanatory view of the burner in which arrangement of the feed hopper of a carbon content compound and oxygen content gas was shown, respectively.

[Drawing 2] (A) and (B) are the whole manufacture furnace outline sectional view of the fullerene concerning the gestalt of operation of the 2nd of this invention, and the explanatory view of the burner in which arrangement of the feed hopper of a carbon content compound and oxygen content gas was shown, respectively.

### [Description of Notations]

The manufacture furnace of fullerene, 11:manufacture furnace body, 12:burner, 13 : 10: The side-attachment-wall section, 14 : An exhaust port, 15:edge wall, 16:refractories, 17:base, 18, the feed hopper of 19:carbon content compound, The feed hopper of the feed hopper of 20 and 21:oxygen content gas, the manufacture furnace of 22:fullerene, 23:manufacture furnace body, 24:burner, 25:side-attachment-wall section, 26:exhaust port, 27:edge wall, 28, 29:entrainment tubing, 30:base, 31, and 32:carbon content compound, 33: The feed hopper of oxygen content gas

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[Translation done.]

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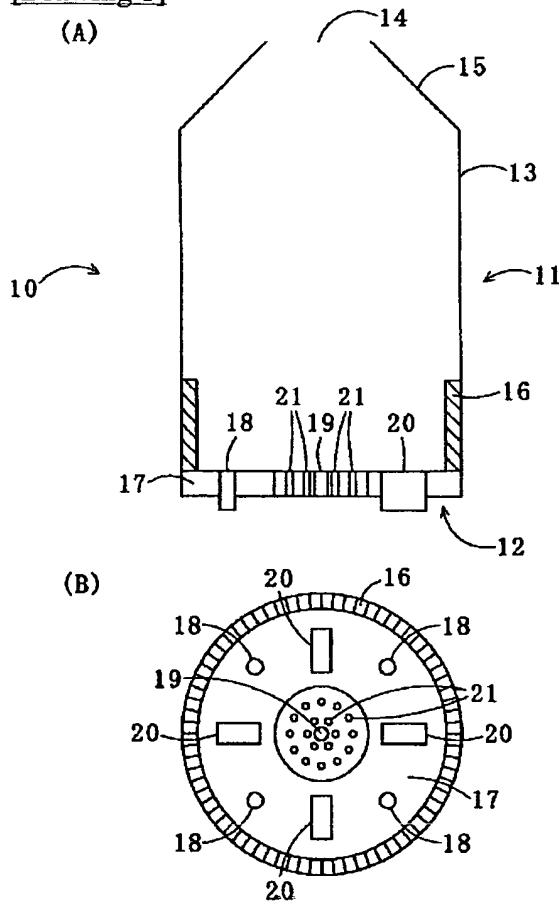
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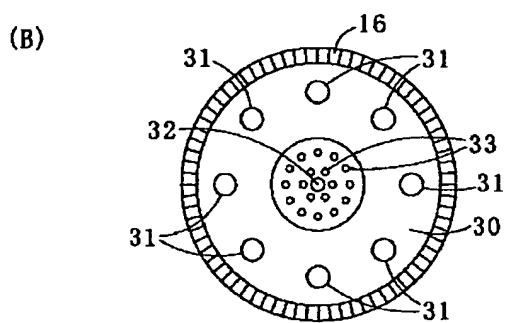
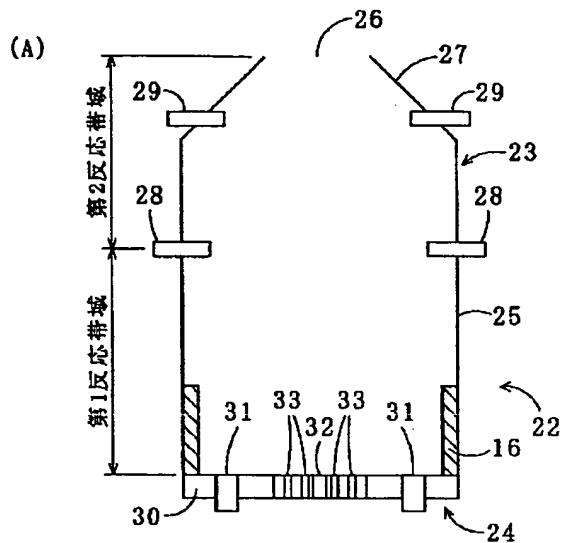
DRAWINGS

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[Drawing 1]



[Drawing 2]



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[Translation done.]

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